

Carbonene Fibers: Toward Next-Generation Fiber Materials

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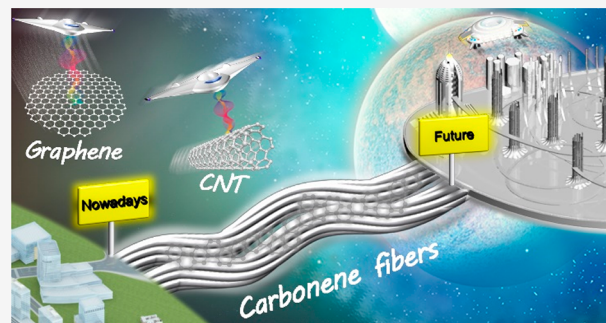
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ABSTRACT: The development of human society has set unprecedented demands for advanced fiber materials, such as lightweight and high-performance fibers for reinforcement of composite materials in frontier fields and functional and intelligent fibers in wearable electronics. Carbonene materials composed of sp^2 -hybridized carbon atoms have been demonstrated to be ideal building blocks for advanced fiber materials, which are referred to as carbonene fibers. Carbonene fibers that generally include pristine carbonene fibers, composite carbonene fibers, and carbonene-modified fibers hold great promise in transferring the extraordinary properties of nanoscale carbonene materials to macroscopic applications. Herein, we give a comprehensive discussion on the conception, classification, and design strategies of carbonene fibers and then summarize recent progress regarding the preparations and applications of carbonene fibers. Finally, we provide insights into developing lightweight, high-performance, functional, and intelligent carbonene fibers for next-generation fiber materials in the near future.

KEYWORDS: carbonene fibers, carbon nanotube, graphene, preparation, application



Starting with natural fibers and moving through the synthetic fiber era to high-performance fibers, fiber materials have achieved great developments along with human society. Especially, the last half-century has witnessed the rapid development of high-performance fibers such as carbon fibers (CFs), aramid fibers, and ceramic fibers, which have been widely used in aerospace, defense, transportation, and other frontier fields. Currently, humanity is exploring greater ocean depths and more of the universe, and the development of a highly efficient, environmentally friendly, and intelligent society have put forward the unprecedented need for advanced fiber materials. Thus, numerous attempts have been made to improve the properties of current high-performance fiber materials as well as develop new-type fiber materials with function and intelligence. Notably, the emergence of nanocarbon materials has significantly promoted the innovation of traditional fiber materials,^{1,2} gradually leading to the post-carbon-fiber era.

Among diverse nanocarbon materials, those materials composed of sp^2 -hybridized carbon atoms show extraordinary electrical, mechanical, thermal, and optical properties (Figure 1a). For example, carbon nanotubes (CNTs) and graphene are representative materials due to their extreme structures in which benzene units are arranged in nanoscale tube/sheet patterns. The ideal CNT and graphene exhibit ultrahigh tensile strength (100–130 GPa, 100 times stronger than steel), Young's modulus (>1 TPa), and specific surface area ($>10^3$ m² g⁻¹) and extraordinary thermal conductivity (3000–6000 W

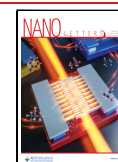
m⁻¹ K⁻¹, 10 times higher than that of copper). The carrier mobility of graphene is higher than 10⁵ cm² V⁻¹ s⁻¹, while CNTs can be metallic or semiconducting depending on their chirality and diameter.^{3,4} To gain an overall grasp, understanding, and utilization of nanocarbon materials with such specific chemical structures, we define the low-dimensional all-carbon materials consisting of sp^2 -hybridized six-membered rings as “carbonene materials” (Figure 1a). For decades, researchers have devoted tremendous efforts to bridge the huge gap between nanoscale structures and macroscopic assemblies of carbonene materials for advanced engineering applications. One significant achievement is the productive development of macroscopic fiber materials based on CNT/graphene, namely “carbonene fibers”, in which the splendid properties of carbonene materials have been favorably accessed. The concept is first stated here to provide a way to discuss these new-type fiber materials: that is, to focus on the common regularity of the design strategy, preparation, and application.

According to the compositions and structures, carbonene fibers can be rationally classified into three types: pristine

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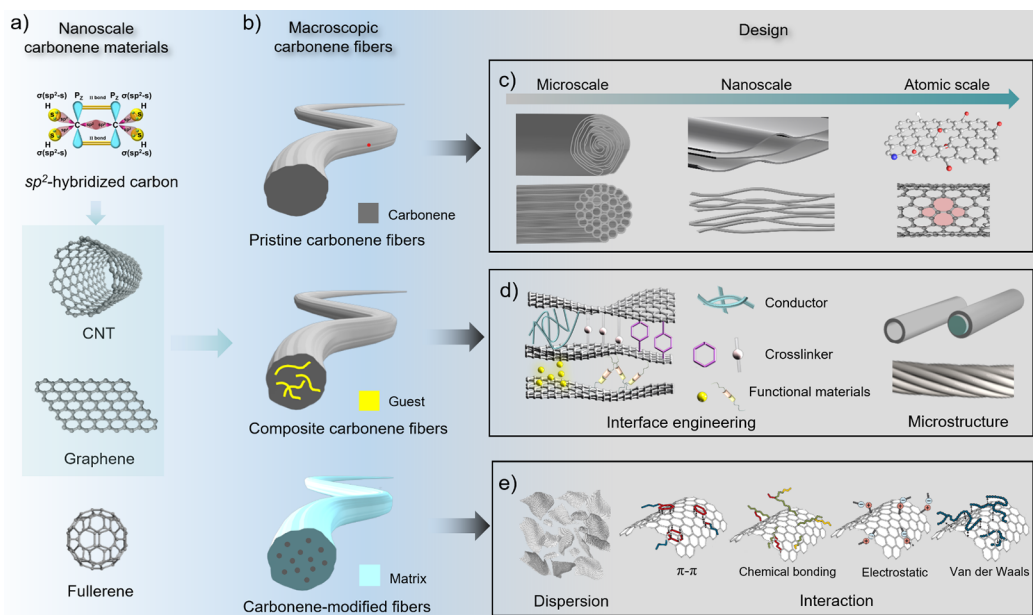


Figure 1. (a) Schematic illustration of nanoscale carbonene materials consisting of sp^2 -hybridized carbon atoms, mainly including CNT, graphene, and fullerene. (b) Three kinds of macroscopic carbonene fibers based on CNT/graphene: pristine carbonene fibers (carbonene materials are the sole building blocks), composite carbonene fibers (carbonene materials are the primary components composited with other guest materials) and carbonene-modified fibers (carbonene materials are additives to other matrix materials). (c) Multiscale design strategies of pristine carbonene fibers on the atomic scale (structural integrity of carbonene materials), nanoscale (closely packed structures, including CNT bundles and graphene laminates), and microscale (homogeneous fiber structure) levels. (d) Design strategies of composite carbonene fibers. The interface engineering between the guest material (conductor, cross-linker, and functional materials) and carbonene materials and the microstructure (twisted or holey structures, for instance) of fibers are two main issues. (e) Design strategies of composite carbonene fibers including dispersion of carbonene materials and their interaction with a matrix. Reproduced with permission from ref 51. Copyright 2018 AAAS.

carbonene fibers, composite carbonene fibers, and carbonene-modified fibers (Figure 1b). Crucial inventions of assembling and processing technologies of carbonene materials are responsible for their blossoming. Pristine carbonene fibers are one-dimensional macroscopic assemblies of pure carbonene materials, typically comprising CNT fibers (CNTFs), graphene fibers (GFs), and CNT/graphene composite fibers (CNTGFs). In composite carbonene fibers, carbonene materials are the primary components combined with other guest materials, which are elaborately introduced for specific applications. In contrast to the structure of composite carbonene fibers, carbonene materials are regarded as the reinforcement or functional component for other fiber materials in carbonene-modified fibers. Because of the intrinsic light weight, ultrahigh mechanical properties, electrical properties, and thermal conductivity and other advantageous characteristics of carbonene materials, carbonene fibers have been demonstrated to possess overall superiority over other traditional fiber materials. For instance, the available CNTFs are on a par with or better than CFs in terms of tensile strength, and they are particularly able to withstand larger deformation (>5%) in comparison to CFs (<2%) and thus can serve as damage-tolerant and ultrastrong fibers with high toughness.^{5,6} The combination of high strength and toughness of carbonene fibers originating from the unique hierarchical structures is inspiring, as those two mechanical parameters are mutually exclusive in traditional materials. Integration of the structures and functions is expected to be realized in carbonene fibers. A telling example is a quadcopter frame composed of CNT yarns, where CNT yarns act as reinforcements and electrical conductors for carrying current to the motors simultaneously.⁷ Lightweight and ultrastrong carbonene fibers

with high conductivity and stability under extreme conditions are potential substitutes for copper wires or high-performance organic fibers. They can serve as multifunctional fibers in aircrafts, submarines, and wind turbines where light weight, superb mechanical properties, and functionalities such as high conductivity, electromagnetic shielding, and radiation and heat resistance are urgently required. In addition, carbonene fibers have been applied as intelligent fibers that can sense and respond to stimuli, which greatly promote the development of artificial muscles and smart systems.^{2,8,9} To summarize, lightweight, high-performance carbonene fibers with function and intelligence are the most promising candidates for next-generation fiber materials.

In this mini review, the concept of carbonene fibers is stated first. Design strategies, preparation methods, and state of the art applications of carbonene fibers are introduced to give a comprehensive understanding of carbonene fibers. Finally, the prospects of carbonene fibers are discussed, which indicates that carbonene fibers would become game-changing fiber materials if their extraordinary potential and multifunctionality can be fully utilized.

A deep understanding of the relationship between the structures and properties of carbonene fibers has been gained through decades of studies, which is essential in guiding the design of carbonene fibers. On account of the distinct structures, the optimal design strategies are introduced with respect to different carbonene fibers, which include pristine carbonene fibers, composite carbonene fibers, and carbonene-modified fibers.

For pristine carbonene fibers, extensive studies have focused on the mechanical, electrical, and thermal performances, with the aim of fabricating new materials to surpass CFs. However,

in spite of the extraordinary properties of carbonene materials on the nanoscale, material defects resulting from the complex hierarchical structures of macroscopic fibers lead to a remarkable degradation of properties across multiple scales. A conventional example is that the tensile strength of SWNTs is predicted to be over 100 GPa, whereas the strongest CNTFs merely possess an average strength of 9.6 GPa.¹⁰ Thus, a multiscale theoretical framework and structural engineering protocol have been established to address this issue and transfer the intrinsic outstanding performance of carbonene materials from nanostructures to macroscopic fibers (Figure 1c).^{11,12}

First, the structural integrity of a carbonene material is the basic requirement for designing pristine carbonene fibers. Controlled synthesis and post treatment of carbonene materials are essential to optimize their atomic structures by reducing structural defects in the sp^2 hexagonal carbon lattice.^{12–14} Except for the primary level structure of individual carbonene materials, second-level (nanoscale) structures of pristine carbonene fibers are closely packed carbonene structures (CNT bundles or graphene laminates). The structures of carbonene building blocks (lengths, diameters, and wall numbers of CNTs or lateral sizes and layer numbers of graphene) and their stacked structures in CNT bundles or graphene laminates are two critical factors of the second-level structure.^{13,15–17} Since the interfacial stress and electron transfer properties between carbonene materials are far inferior to those of individual carbonene materials in most cases, the fracture mechanism of pristine carbonene fibers can be simplified to the sliding displacement between CNTs or graphene layers, and the electrical conductivity is dominated by the intertubular and interlayer transport of carriers.^{18,19} Sufficient interaction between the adjacent carbonene building blocks are in favor of the mechanics and transport properties of these assemblies. Meanwhile, a lower boundary of building blocks is required to minimize the boundary-related defect densities, which leads to poor stress transfer and scattering of electrons and phonons. Consequently, high-aspect-ratio CNTs or large graphene nanosheets with perfect and compact alignment in bundles or laminates can help to obtain sufficient interactions and overlap between individual carbonene building blocks, so that effective stress transfer and fast transport of electrons successfully occur in the aggregates.^{17,20} For example, CNT bundles with centimeter-long lengths have been rationally designed to release the different initial strains and achieve a uniform alignment of CNTs, exhibiting a tensile strength of over 80 GPa, which is comparable to that of individual CNTs.²¹ At a third level (microscale), a homogeneous structure over the entire fibers with a high degree of orientation and densification of CNT bundles or graphene laminates is needed to eliminate microscale voids and an entangled topology and to flatten wavy morphologies.^{12,22} Moreover, impurities such as residual metal catalysts in CNTs are regarded as the defect points in pristine carbonene fibers and account for the reduction of their mechanical performance. Purification of carbonene materials is required before the assembly process.¹⁴ In particular, the thermal conductivity of GFs has an advantage over that of CNTFs due to their large-sized crystalline domains for efficient phonon transport, which also indicates a practical design to obtain high heat conduction of carbonene fibers by constructing highly crystalline structures.²⁰ The mutual interaction between CNT and graphene and the comple-

mentary enhancement of the connectivity of carbonene networks are attractive for improving the torsional strength and electrical conductivity of CNTGFs.^{23–28}

As was discussed above, the interfacial properties of carbonene building blocks highly determine the performances of macroscopic fibers. Modulation of second- and third-level structures is prominent in the design of pristine carbonene fibers. In practice, researchers focus on the alignment and stacking structures of carbonene materials, which can be readily modulated by stretching and densification techniques. However, the merit of ultralong CNTs and large graphene nanosheets has not been fully utilized because of the difficulty in the assembly and processing. The design and processing of well-aligned structures of ultralong CNTs and large graphene nanosheets in fibers need dedicated efforts in the future.

In composite carbonene fibers, carbonene materials are combined with other guest materials such as polymers, metals, small molecules, and other nanomaterials for diverse applications.^{29–34} The characteristics of additive materials and their interaction with carbonene materials are of fundamental importance in the design of composite carbonene fibers (Figure 1d). In order to improve the efficiency of stress transfer in intertubular or interlayer structures of CNTs or graphene nanosheets, additive materials that are able to cross-link carbonene materials through covalent bond, hydrogen bond, π - π van der Waals, or synergistic interactions are introduced. Remarkably, some flexible polymer chains such as poly(vinyl alcohol) (PVA) or monomers of resins can act as plasticizers to promote the orientation and alignment of CNTs under tension, as well as to fill the micro voids and thus densify the fibers. Those effects of a guest material can synergistically improve the mechanical properties of the composite fibers.^{35,36} However, the introduction of an insulating guest material would hinder the electron transport between individual building blocks because of the enlarged interfacial resistance.^{32,37} This strategy is unfavorable for electrical conductivity. Thus, conducting materials such as conducting polymers or metals are commonly used to bridge the intertubular or interlayer conductive paths.^{29,38–40} Doping of fibers is another way to improve electrical conductivity by increasing the carrier density of composite carbonene fibers, while their durability in an ambient environment is of great concern, since most of them are unstable in air.⁴¹

So far, cross-linking and densification have been the most efficient strategies for making strong composite carbonene fibers. A strong interaction derived from covalent cross-linking is preferred in comparison with other weak noncovalent interactions.^{32,42–44} However, the introduction of active cross-linking sites on carbonene materials may sacrifice their intrinsic properties, which results in a disadvantaged structure at the atomic level. In most cross-linking models, the interaction occurs between the tube walls of CNT or planes of graphene.^{29,32,40,42} Attention may be given to building a strong interaction of carbonene materials through covalent cross-linking at ports of CNTs and edges of graphene nanosheets. This strategy would be an effective way to solve the dilemma that covalent cross-linking of carbonene is at the cost of the properties of an individual tube or nanosheet.

Integrated superior properties of carbonene materials, such as intrinsic flexibility, large surface area, high mechanical properties, electrical/thermal conductivity, and chemical stability, can be utilized to design functional and intelligent fiber materials by incorporating guest materials. Species of

functional guest materials and their interaction with carbonene materials, together with morphologies and microstructures of the frameworks assembled from carbonene materials, are the key elements in the design of those fibers (Figure 1d). For instance, fibers with twisted and coiled architectures can be used as a powerful torsional and tensile artificial muscle, in which the untwisting of structures can transfer a radial volume expansion to a rotational actuation or axial contraction.⁸ After introduction of guest materials with large thermal expansion into fibers, the actuation of composite carbonene fibers would be generated by electrical or light stimuli because of the photothermal and electrothermal conversion ability of carbonene materials.³⁴ Porous CNT and graphene fibers are ideal conductive frameworks with ultrahigh specific surface areas for loading guest materials. By the compositing of electrochemically active materials, fiber-shaped electrodes are readily obtained and can be applied to construct wearable energy conversion and storage devices.^{1,30,45} The rational design of hollow, core–shell, or sandwiched composite structures of fibers can realize a two-electrode capacitor system in a single fiber. Their interfacial resistance can be reduced by a stable interface and increased contact surface between the electrolyte and the electrodes. Due to the integrated superiorities of fiber-shaped frameworks assembled from carbonene materials, multifunctional and intelligent composite carbonene fibers are accessible from ingenious design of the hierarchical composite structures.¹

One efficient design of carbonene-modified fibers is constructing a composite structure by their acting as fillers.^{46–48} The structure and dispersion of carbonene materials, along with interfacial properties between fillers and matrices, are major issues that need careful consideration (Figure 1e). First, the extraordinary properties of carbonene materials are the prerequisites of their functions in fiber matrices. However, utilization of carbonene materials with an intact structure is a challenge due to the issues of dispersion and interfacial properties. This concern stems from the adverse agglomeration of carbonene materials and weak interfacial interaction in fibers. The presence of van der Waals forces between carbonene materials causes their tendency to agglomerate in the matrices; meanwhile, strong filler–matrix bonds are difficult to obtain due to their atomically smooth surfaces. Noncovalent functionalization of carbonene materials is helpful in the dispersion but fails to introduce strong interfacial interactions with matrices. Chemical functionalization is commonly used to improve the dispersion of carbonene materials and provide ample interaction with matrices, but it would damage their intrinsic properties. Thus, the strategy is to design an optimal decoration of carbonene materials to find a balance between an enhanced interface and reduced intralayer or intratube properties.⁴⁹ Precise functionalization of CNT ports or graphene nanosheet edges is another effective strategy but is also a challenge. Notably, carbonene materials can induce a superior structure of the matrix. For instance, polymer chains in the vicinity of carbonene materials have been observed to exhibit more compact packing and better orientation in comparison to a bulk polymer. The existence of interphase polymers in composite fibers can increase their performance.⁵⁰ The structural evolution of fiber matrices with carbonene material fillers should be particularly probed to reveal the enhancement mechanism comprehensively.

The discussion of the structure and dispersion of carbonene materials also refers to their morphologies and arranged

networks in fiber matrices. It is worth noting that CNT and graphene are quite distinct in geometrical structure. Thus, the employment of CNT or graphene in composite fibers requires careful thought. The nature of the interface between matrices and carbonene materials is jointly controlled by their structural characteristics (e.g., 1D or 2D shape, length, or aspect ratio) and interaction with adjacent matrices, which highly determines the stress transfer in composite fibers under mechanical deformations. The effect of CNTs on the mechanical properties of composite fibers has been demonstrated to rely more on tube orientation in comparison to graphene nanosheets, and randomly oriented graphene nanosheets are superior to randomly distributed CNTs in reinforcement.⁵¹ For the electrical properties of carbonene-modified fibers, CNTs have a pronounced effect in improving the performance relative to graphene, since 1D nanotubes are easier to interlace with each other into a network through overlapping contact in comparison to 2D nanosheets at a low filler concentration. For 2D nanosheets, their wrinkle and crimple structures result in a low effective aspect ratio and only a plane-to-plane contact between them is helpful to construct a conductive pathway.⁵² The adoption of long CNTs or large graphene nanosheets is theoretically preferred, but carbonene materials with small dimensions are still commonly used due to the dispersion-related problems in practice. Because of their intrinsic flexibility, carbonene materials tend to exhibit crumpled and folded configuration in matrices, leading to an insufficient exposure of the surface area to interact with adjacent matrices or a failure to connect with each other. Therefore, carbonene materials with an extended structure and high alignment along the axial direction of fibers are in demand when one aims to produce fiber materials with a high tensile strength and modulus and obtain a percolated conductive network at a low concentration of carbonene materials. Notably, applications of two or more kinds carbonene materials in a hierarchical structure have been found to show a synergistic enhancement in composite performance.⁵³ Another composite structure is where carbonene materials are coated on the surface of fibers. On the one hand, a conformal coating of a compact carbonene layer on fiber matrices can enhance the mechanical properties and regulate the surface properties of fiber matrices. Hydrophilic and hydrophobic performances of a composite can be tailored by carbonene materials with diverse chemical structures, while functions can be introduced in composite fibers due to the electrical and thermal conductivity of carbonene materials.⁵⁴ On the other hand, composite fibers coated with vertical or porous carbonene material layers are common electrodes for sensors, supercapacitors, and catalyst supports.⁵⁵ The interface interaction needs careful evaluation and thought in order to gain good stability and durability of the composite fibers under operation.

To bridge the gap between nanoscale carbonene materials and their macroscopic applications, a controlled synthesis of carbonene materials and a practical preparation of carbonene fibers are required to obtain the expected structures with specific properties. Tremendous attempts have been devoted to preparing high-performance carbonene fibers not only for experimental verification but also for going beyond the laboratory to industry. The representative preparation and processing methods are presented here together with a discussion of the properties to give an overall introduction to the vital issues involved in the carbonene fiber field.

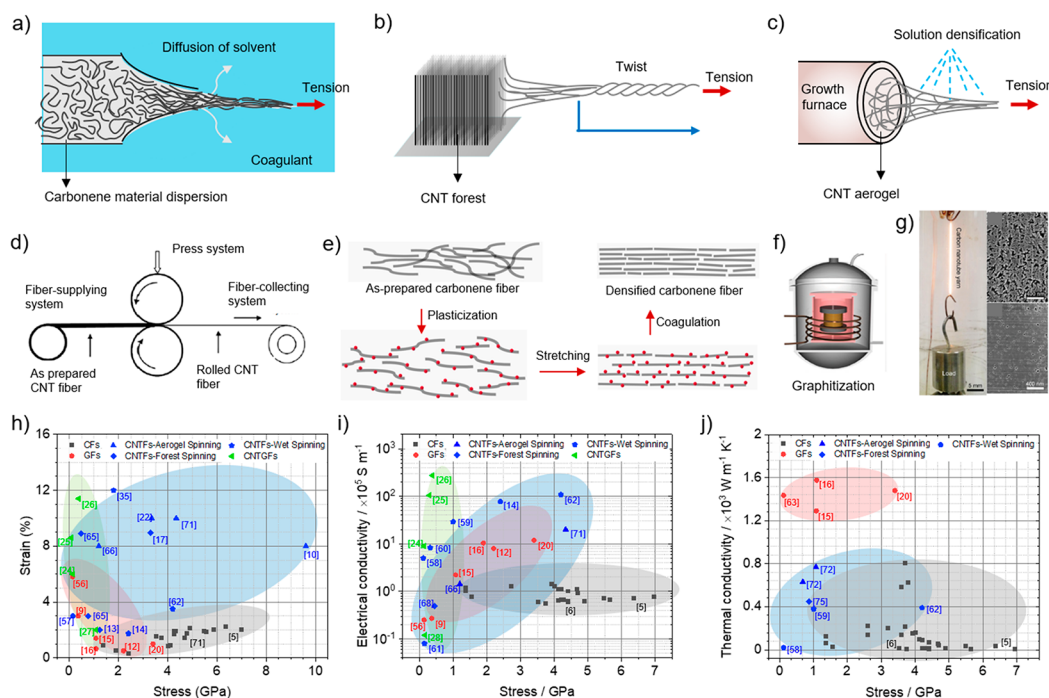


Figure 2. Schematic illustrations of (a) wet-spinning, (b) forest-spinning, and (c) aerogel-spinning procedures of pristine carbonene fibers. (d) Schematic of the system for mechanical rolling of CNTFs. Reproduced from ref 71 with permission. Copyright 2014 Nature Publishing Group. (e) Schematic illustration of the plasticization-assisted stretching of pristine carbonene fibers. Reproduced from ref 12 with permission. Copyright 2016 Wiley-VCH. (f) Graphitization of GFs in a graphitization furnace. Reproduced from ref 74 with permission. Copyright 2016 Wiley-VCH. (g) Setup for applying an incandescent tension annealing process to CNTFs via Joule heating (left) and the cross-sectional SEM images before (upper right) and after (bottom right) treatment. Reproduced from ref 74 with permission. Copyright 2016 Wiley-VCH. (h–j) Comparison between CFs and pristine carbonene fibers on tensile stress, strain, and electrical and thermal conductivity. Black ■, red ●, blue ▲, blue ◆, blue ⬡, and green ◀ represent the data of CFs, pristine GFs, CNTFs from aerogel, forest, and wet spinning, and CNTGFs, respectively.

Wet spinning is a general technique for manufacturing commercial synthetic polymer fibers, which has also been proved to be suitable for the preparation of carbonene fibers. In the process of wet spinning, carbonene materials or a derivative such as graphene oxide (GO) are first dispersed into solution and injected into a coagulation bath.^{56,57} The resultant pristine carbonene fibers are formed after double diffusion between the solution and coagulation and finally collected by continuous withdrawal (Figure 2a). The synthesis of homogeneous, stable dispersions of carbonene materials is a critical step in wet spinning. To overcome the entangled and stacked aggregating of CNTs, fuming sulfuric acid or chlorosulfonic acid is adopted to disperse CNTs due to the strong protonation effect.^{58–60} Conversely, GO is a common precursor of GFs because of its good dispersibility resulting from repulsion interactions between oxygen-containing groups on the basal plane. The merit of wet spinning is that pristine carbonene fibers with high performance can be feasibly fabricated by the employment of carbonene materials with advantageous structures.^{14,61} Meanwhile, liquid-crystal spinning based on the dispersion with a liquid phase is promising in achieving carbonene materials with high orientation and good alignment.^{59,62,63} Liquid-crystal spinning of CNTs with a high purity and aspect ratio (12 μm length and 1.76 nm diameter) could fabricate CNTFs with a high tensile strength of 4.2 GPa and an electrical conductivity of up to 10.9 MS m^{-1} .⁶² Forest and aerogel spinnings are regarded as dry spinning methods in contrast to wet spinning.^{64–67} In forest spinning, the CNTFs are continuously drawn from vertically aligned CNT forests, while in aerogel spinning, CNTFs are

directly collected from CNT aerogels from a growth furnace in which floating catalyst chemical vapor deposition (CVD) is conducted (Figure 2b,c). As structures of assemblies just drawn from dry-spinning methods are noncompact, twisting and solution densification are widely introduced before the collection of fibers.^{22,68,69} In comparison to wet spinning, in dry spinning ultralong CNTs (hundreds of micrometers to millimeters) are readily assembled into fiber without dispersion.^{17,70} For example, spinning a CNT forest with 1 mm height could give a CNTF with a tensile strength of up to 3.3 GPa.⁷⁰ Forest spinning is an effective technique to fabricate CNTFs with favorable alignment and high purity due to the vertical nature of CNT forests and the remaining catalysts on substrates, but its industrial application is hindered by high cost and difficulties in mass fabrication. Aerogel spinning is compatible with industrial production, but the strongly entangled, inhomogeneous, and impurity-embedded structures of CNTs remain crucial problems in fabricating CNTFs with a high alignment and compact structure. The spinning methods are still rough techniques because of the difficulties in the controllable assembly of advantageous second- and third-level structures in fibers. This situation calls for the development of post treatments to optimize the hierarchical structures of as-prepared pristine carbonene fibers.

Solution densification driven by a capillary force from the evaporation of polar solvents and mechanical densification conducted under external force are effective ways to acquire a dense structure.^{69,71,72} Roller pressing can boost the tensile strength of CNT assemblies to 5.53–9.6 GPa (Figure 2d).^{2,10,71} However, the alignments of carbonene materials

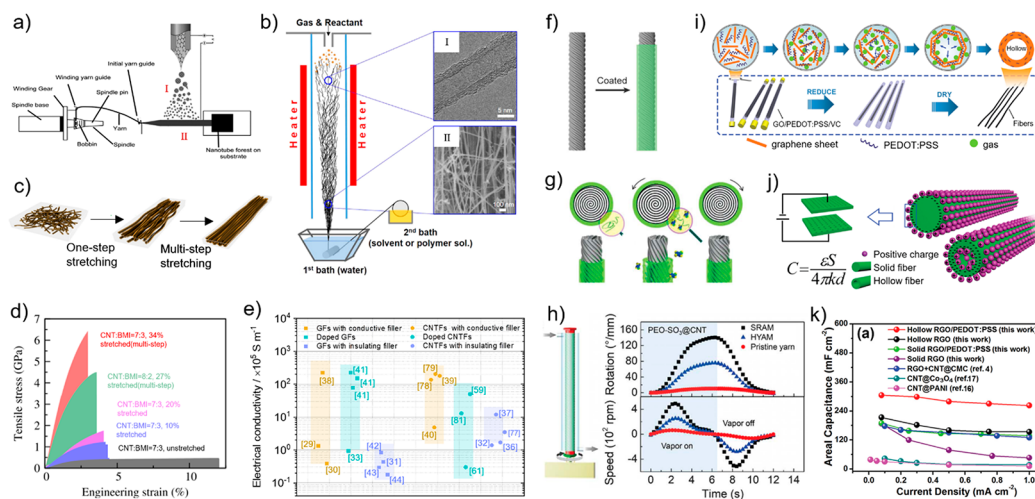


Figure 3. (a) Setup for continuously producing composite CNT yarns by spraying (I) a guest material dispersion onto (II) a CNT sheet drawn from a spinnable forest. Reproduced from ref 24 with permission. Copyright 2014 Wiley-VCH. (b) Schematic illustration of incorporating a polymer in CNTFs during an aerogel spinning-process and (I) TEM image of the synthesized CNTs and (II) SEM image of the CNT assemblies with a loose structure. Reproduced from ref 77 with permission. Copyright 2015 Elsevier BV. (c) Illustration of the structural evolution of CNT/BMI composite fibers under stretching. Reproduced from ref 36 with permission. Copyright 2015 Nature Publishing Group, licensed under a Creative Commons Attribution (CC BY) license: <http://creativecommons.org/licenses/by/4.0/>. We have moved “Resin-impregnation” and the schematic structure of CNT without resin and rearranged the position of schematic structures of composite film after one-step stretching and multistep stretching from the original label position. (d) Typical stress–strain curves for CNT/BMI composites prepared by different stretching processes. Reproduced from ref 36 with permission. Copyright 2015 Nature Publishing Group, licensed under a Creative Commons Attribution (CC BY) license: <http://creativecommons.org/licenses/by/4.0/>. No changes were made. (e) Electrical conductivity of composite carbonene fibers. (f) Schematic of a twisted CNT yarn coating with a polymer sheath. (g) Illustrations (left to right) of a sheath-run artificial muscle (SRAM) before vapor exposure and during vapor sorption and then desorption, which caused yarn untwist and uptwist, respectively. (h) Illustration of vapor delivery to a muscle and plots of the time dependence of the torsional stroke and rotation speed for a SRAM, a hybrid yarn artificial muscle (HYAM), and a pristine CNT yarn. Reproduced from ref 85 with permission. Copyright 2019 AAAS. (i) Schematic illustration of the preparation of hollow fibers. (j) Illustration of the charge distribution on a solid fiber, hollow fiber, and parallel-plate capacitor. (k) Electrochemical performances of the fiber supercapacitors. Reproduced from ref 45 with permission. Copyright 2016 Wiley-VCH.

are markedly limited by their randomly entangled or stacked structures and intertubular or interlayer cross-linking sites. Drawing during the formation process of fibers helps to densify their microstructure and simultaneously arrange the building blocks to a certain extent. Stretching fibers with enhanced deformable plasticity by plasticizers (e.g., acetic acid for GO nanosheets and chlorosulfonic acid for CNTs) can generate highly compact and aligned microstructures (Figure 2e).^{20,73} The treatment of pristine carbonene materials under extreme conditions is another effective postprocessing method. Extreme-high-temperature annealing (Figure 2f) is a requisite procedure for the transformation of GO fibers to GF in which the atomic-scale defects of nanosheets are restored and graphitization domains with densified structures are generated. The transformation contributes to a synchronous enhancement of the mechanical, electrical, and thermal properties. Xu et al. reported highly crystalline GFs with a tensile strength of 3.4 GPa, an electrical conductivity of $1.19 \times 10^6 \text{ S m}^{-1}$, and a thermal conductivity of $1480 \text{ W m}^{-1} \text{ K}^{-1}$ by combining a liquid-crystal spinning method with a plasticization spinning process and a thermal treatment at $2800 \text{ }^\circ\text{C}$.²⁰ For CNTFs, electron/ion beam irradiation or Joule heating treatment of fibers under tension can produce intertubular cross-linking and densify the microstructure (Figure 2h).⁷⁴

Carbonene fibers have long been considered as promising competitors to CFs in future commercial applications demanding lightweight, high-strength, and functional fiber materials ever since they were discovered. Here a comprehensive comparison between pristine carbonene fibers (GFs, CNTFs, and CNTGTs) and CFs with regard to the

mechanical performance and electrical and thermal conductivity is presented and evaluated on the basis of the representative reported data (Figure 2f–h). It is clear that pristine carbonene fibers have a great advantage over CFs with regard to tensile strain, and thus they are regarded to be high-performance fibers with particularly light weight, high strength, and high toughness. Pristine carbonene fibers exhibit better flexibility to withstand deformations of higher than 5% and sustain structural integrity when they are bent and knotted, whereas CFs would surrender to deformation under similar conditions. In contrast, CFs are strong and stiff fiber materials with extremely high Young’s moduli (159–960 GPa), while those of pristine carbonene fibers are mainly in the range of 100–400 GPa.^{5,6,71} Apart from their mechanical performances, pristine carbonene fibers possess higher electrical conductivity in comparison to that of CFs, profiting from the sp^2 -hybridization structure of carbon atoms. In particular, pristine carbonene fibers prepared from wet spinning are the most conductive due to their compact and well-aligned structure.⁶² The thermal conductivity of GFs far exceeds that of CNTFs and CFs, which stems from the large-sized graphitized crystals in their structure.^{20,75} In summary, pristine carbonene fibers may be employed as high-performance fiber materials with potential in specific applications requiring high electrical and thermal conductivity.

The preparation process of composite carbonene fibers is compatible with that of pristine carbonene fibers. In wet spinning, adoption of a guest material can be conducted by preparing composite spinning solutions. Mixing a GO aqueous dispersion with a phenolic resin solution followed by wet-

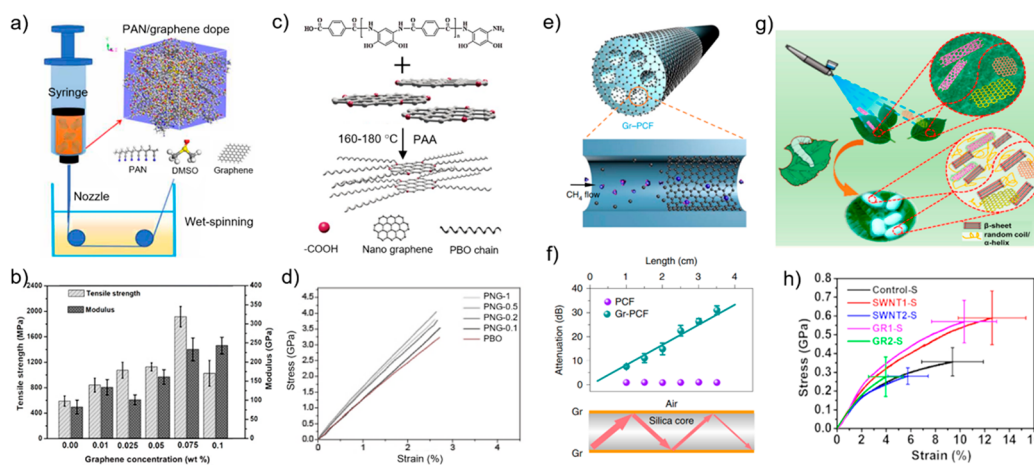


Figure 4. (a) Fabrication of PAN/graphene precursor fibers from a mixture. Adapted from ref 87. Copyright the Authors, some rights reserved; exclusive licensee AAAS. Distributed under a CC BY-NC 4.0 license: <http://creativecommons.org/licenses/by-nc/4.0/>. We have simplified the schematic illustration of the wet-spinning process with one bath reserved and rearranged the position of PAN, DMSO, and graphene from the original label position. (b) Mechanical properties of the carbonized PAN/graphene CFs. Adapted from ref 87. Copyright the Authors, some rights reserved; exclusive licensee AAAS. Distributed under a CC BY-NC 4.0 license: <http://creativecommons.org/licenses/by-nc/4.0/>. No changes were made. (c) Preparation of PNG by *in situ* polymerization. Reproduced from ref 48 with permission. Copyright 2016 MDPI, licensed under a Creative Commons Attribution (CC BY) license: <http://creativecommons.org/licenses/by/4.0/>. We have moved the schematic illustration of reaction monomers and reaction conditions for the preparation of poly(*p*-phenylenebenzobisoxazole) and rearranged the position of PBO and PNG from the original label position. (d) Typical stress–strain curves of PBO and PNG composite fibers with different NGO contents. Reproduced from ref 48 with permission. Copyright 2016 MDPI, licensed under a Creative Commons Attribution (CC BY) license: <http://creativecommons.org/licenses/by/4.0/>. No changes were made. (e) Schematics of CVD method for graphene–photonic crystal fibers. (f) (top) Measured optical attenuation of light propagation with different fiber lengths and (bottom) schematic of light attenuation with multiple reflections during its propagation along the graphene–photonic crystal fiber. Reproduced from ref 89 with permission. Copyright 2019 Nature Publishing Group. (g) Schematic showing the natural process of incorporating SWNTs or graphene into silk by feeding silkworms with mulberry leaves sprayed with the corresponding solutions. (h) Stress–strain curves of degummed silk fibers. Reproduced from ref 47 with permission. Copyright 2016 American Chemical Society.

spinning and annealing can largely increase the tensile strength from 0.68 GPa for GFs to 1.45 GPa for composite GFs by reducing defects and forming new C–C bonds in fibers.⁴² The porous and loose structures of as-drawn CNT assemblies from vertical forests or reaction furnaces provide large surface areas and vacancies for a combination of external materials by coating, infiltration (Figure 3a,b), and so on.^{34,76–79} Composite carbonene fibers with dense structures are obtained after subsequent twisting, biscrolling, evaporation-driven shrinking, stretching, or mechanical pressing.^{34,39,69,73} Impregnating a CNT aggregate with bismaleimide (BMI) enabled the structural relaxation of composite fibers by stretching in a multistep way. After a curing process, CNT/BMI composite fibers with a compact and aligned structure exhibited ultrahigh tensile strengths of up to 6.94 GPa (Figure 3c,d).³⁶ Among the diverse guest materials, incorporation of a conducting polymer or a metal can simultaneously enhance the mechanical and electrical performances.^{29,39,40} The electrical conductivity of composite carbonene fibers is summarized in Figure 3e, which shows that composite fibers with conductive fillers have generally higher conductivity in comparison to fibers composited with insulating guest materials. Doping pristine carbonene fibers with dopants (e.g., K, Br₂, I₂, and FeCl₃) has a prominent effect on optimizing their conductivity (Figure 3e).⁴¹ What is more, Liu et al. doped GFs with calcium, and the resultant intercalated GF exhibited a superconducting transition at ~11 K, approaching that of commercial NbTi conductors.⁸⁰ Electrodeposition of dopants and postcapping method have been exploited to address the operation stability of doped fibers.⁸¹

To explore the application of composite carbonene fibers as functional and smart fibers, preparation strategies such as modified spinning,^{82,83} confined thermal reduction,⁴⁵ and template-assisted growth methods⁸⁴ have been developed to assemble composite carbonene fibers with hierarchical structures. For applications in artificial muscle fiber fields, thermally, electrically, photonically, or chemically powered hybrid CNT yarn muscles have been reported through infiltrating yarns with paraffin or biscrolling yarns with a palladium layer.³⁴ Mu et al. demonstrated that CNT yarn artificial muscles with sheath-run structures show a higher work capacity and lower cost in comparison to CNT yarns filled with guest materials. Thus, the guest materials were coated on a twisted or coiled core of CNT yarns to fabricate high-performance artificial muscles (Figure 3f–h).⁸⁵ An example of the fabrication of fiber supercapacitors based on composite carbonene fibers is that a hollow fiber could be formed during a confined hydrothermal reduction in a well-sealed glass tube (Figure 3i). On account of the increased exposed area (Figure 3j), the hollow composite carbonene fiber render a much higher capacitance (304.5 mF cm², 143.3 F cm⁻³, or 63.1 F g⁻¹) in comparison to that of solid fibers (Figure 3k).⁴⁵

The dispersion of carbonene materials is the most critical issue in the preparation of carbonene-modified fibers in which carbonene materials are expected to be homogeneously dispersed in matrices or coated on the surface. Processing technologies, including physical blending, *in situ* addition, solution coating, and CVD techniques, have been employed to fabricate carbonene-modified fibers in both the laboratory and industry. In physical blending, carbonene materials are

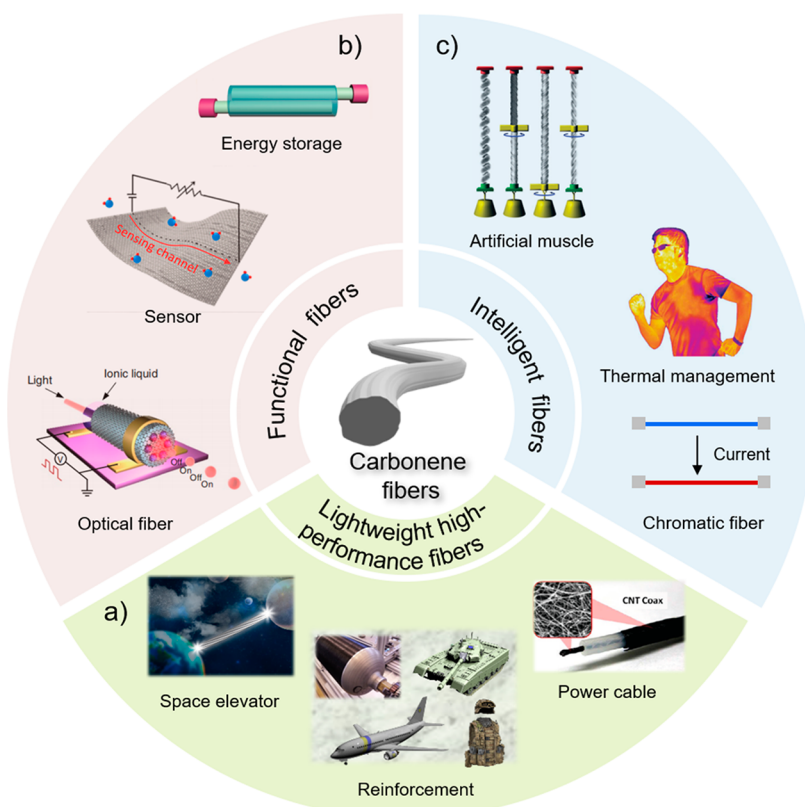


Figure 5. Applications of carbonene fibers toward (a) lightweight and high-performance fibers (power cable, reproduced from ref 92 with permission, copyright 2012 American Chemical Society), (b) functional fibers (energy storage, reproduced from ref 26 with permission, copyright 2015 American Chemical Society, sensors, reproduced from ref 94 with permission, copyright 2019 American Chemical Society, and optical fibers, reproduced from ref 89 with permission, copyright 2019 Nature Publishing Group), and (c) intelligent fibers (artificial muscle, reproduced from ref 34 with permission, copyright 2012 AAAS, and thermal management, reproduced from ref 96 with permission, copyright 2019 AAAS).

chosen to disperse in solvent consistent with that of matrix materials before blending or directly disperse in matrix materials according to the processing features. Particular treatments are carefully adopted by considering the viscosity-related issue during operation. Mechanical stirring and sonication are usually used to pretreat a dilute carbonene dispersion, while intense shear from ball milling and calendaring methods is needed in processing a high-viscosity mixture of carbonene and matrix materials.⁸⁶ After dispersion of carbonene materials, composites are subsequently processed to form fibers. Blending the stable graphene nanosheet/dimethyl sulfoxide dispersion with a PAN polymer followed by the preparation procedures of CFs can largely increase the tensile strength and modulus of CFs from 589 and 82 GPa to 1916 and 233 GPa, respectively (Figure 4a,b).⁸⁷ Chemical functionalization of carbonene materials has been carried out before blending to overcome dispersion challenges and enhance the filler–matrix interactions, such as hydrogen-bonding or electrostatic interactions. Shear mixing of functionalized graphene containing pendant hydroxyl groups with poly(methyl methacrylate) (PMMA) can prepare graphene-modified fibers with enhanced mechanical and thermal properties, in which hydrogen bonds are formed between the hydroxyl groups of functionalized graphene and the carbonyl groups of PMMA.⁴⁶

In situ addition refers to compositing carbonene materials in the synthesis procedure of matrix materials, which is widely applied to the preparation of carbonene-modified polymer materials. In comparison with physical blending, *in situ*

polymerization can readily solve the viscosity-related problem, as the initial state of the composite system before polymerization exhibits a low viscosity. In addition, *in situ* polymerization shows sufficient potential in tailoring the interface properties. By decoration of carbonene materials with functional groups to react with monomers, the carbonene material can participate in the polymerization as a “special monomer”. The formation of a covalent bond can significantly improve the interfacial interaction and promote stress transfer. For example, poly(*p*-phenylenebenzobisoxazole) (PBO)/nanographene (NG) composites (PNG) were prepared via *in situ* polymerization, accompanied by the reduction of nano graphene oxide to NG. The composite fiber with 0.5 wt % NG exhibits 25% and 41% increments in tensile strength and modulus in comparison with neat PBO fibers, respectively (Figure 4c,d).⁴⁸ Grafting graphene nanosheets with nylon-6 chains can enhance the tensile strength of graphene-modified nylon-6 fibers by 2.1-fold with a graphene loading of 0.1 wt %.⁸⁸ Although research has been carried out for decades in this field, the individual dispersion of carbonene materials is still difficult. In comparison with searching for new combinations of carbonene and other materials, a deep understanding and development of dispersion as well as composite techniques will be more important for full utilization of carbonene materials in the future.

Coating of carbonene materials onto other fiber-shaped materials is a simple way to prepared a hierarchical structure of carbonene-modified fibers. Relative to solution dipping–coating methods, the direct growth of carbonene materials

on the surface of fibers eliminates the dispersion procedure and is able to precisely regulate the thickness of the coating layer with ease. Chen et al. successfully deposited a uniform graphene layer with an average thickness of ~ 2.0 nm on the inside wall of a porous photonic crystal optical fiber through CVD and realized a greatly enhanced light–matter interaction between graphene and core-guided light (Figure 4e,f).⁸⁹ Beyond previous techniques, unique preparation methods are also under continuous development. Typically, by creative feeding of *Bombyx mori* larval silkworms with SWNTs or graphene, carbonene-modified silk fibers with enhanced mechanical properties can be directly collected (Figure 4g,h). The elongation at break and modulus of composite fibers are increased because the carbonene materials can impede the conformation transition of a silk fibroin from a random coil and α -helix to a β -sheet.⁴⁷

Recent decades have witnessed remarkable achievements in carbonene fiber fields, and the academic community has made numerous attempts to find their killer applications. The overall applications of carbonene fibers can be classified into three aspects: lightweight and high-performance fibers, functional fibers, and intelligent fibers (Figure 5).

Carbonene fibers with light weight and outstanding mechanical performance are expected to be applied in industries of civilian areas or frontier fields such as aerospace, military, and defense. For instance, carbonene fibers can serve not only as a strong fabric for individual protection but also as lightweight reinforcements for structural materials in automobiles and airplanes and even in aircraft and submarines, where the load-carrying capacity under extreme conditions is urgently needed. An experimental result demonstrated that overwrapping aluminum rings with thermoset/CNT yarn can significantly increase the room-temperature breaking load by over 200% relative to bare Al rings, while merely leading to an 11% increase in weight. It provided a strategy to fabricate lightweight composite overwrapped pressure vessels.⁹⁰ It is fascinating that CNTs appear to be candidates that meet the strength to mass ratio requirement ($48.5 \text{ GPa g}^{-1} \text{ cm}^{-3}$) for constructing a space elevator, and a blueprint for its adventurous future application has been drawn.⁹¹ Moreover, although the reported conductivities of carbonene fibers are still less than that of commercial copper wires ($5.5 \times 10^7 \text{ S m}^{-1}$) (Figure 3e), the advantage of light weight (carbonene fiber density, $< 2 \text{ g cm}^{-3}$; copper wire density, 8.9 g cm^{-3}) endows carbonene fibers with specific conductivity comparable to that of the copper wire ($6.18 \text{ kS m}^2 \text{ kg}^{-1}$), with higher values surpassing that of copper in particular when carbonene fibers are doped (the specific conductivity of GF doped with K 13.8 $\text{kS m}^2 \text{ kg}^{-1}$).^{5,41} Thus, in combination with their high stability and mechanical robustness, highly conductive carbonene fibers have a great potential to replace copper wires as power cables for electrical power transmission. A prototype was presented in which GFs were applied in lightweight rotator coils to assist the operation of an electrical motor with a high speed of ~ 350 rpm at a bias of 8 V.¹² CNT coaxial cables were also reported to be approaching a level of performance that will give them a competitive edge over cables constructed from metallic conductors.⁹² Furthermore, as carbonene materials have an excellent transport ability, lightweight and high-performance carbonene fibers can be equipped with attractive functions such as electromagnetic shielding and infrared stealth, promoting their applications for defense and military fields.

Because of the large specific surface area and unique electrical, thermal, and optical properties of carbonene materials, functional carbonene fibers are widely applied in energy storage and conversion electronics (supercapacitors, batteries and nanogenerators), sensors, optoelectronic devices, neural recording systems, etc.^{89,93–95} The intrinsic mechanical characteristics of carbonene materials ensure the flexibility of these fiber-shaped devices and allow them to be easily integrated with fabrics to produce wearable electronics. For intelligent applications, carbonene fibers are able to conduct actuation, thermal management, and color changing and undergo smart reactions under a stimulus.^{34,96,97} A carbonene-modified fiber textile with a thin layer of CNTs on the surface of triacetate-cellulose bimorph fibers can modulate infrared radiation as the relative humidity of the underlying skin changes. Smart thermal management of the human body can be realized by a dynamic gating effect derived from electromagnetic coupling between adjacent fibers.⁹⁶ The rise of the Internet of Things, artificial intelligence, comprehensive health, and other advanced fields demands smart systems assembled with functional and intelligent elements, including energy-harvesting and storage devices, sensors, actuators, and so on. Particularly, those systems can be rationally designed by integrating multifunctional and intelligent carbonene fibers.

Due to their extraordinary properties, research on carbonene materials has lasted for decades and is still growing at present. The successful assemblies of nanoscale carbonene materials into macroscopic fiber materials and their applications in plentiful areas has been carefully thought out, as introduced in this mini review. Carbonene fibers are considered to be fiber materials composed of carbonene materials in which the intrinsic properties of carbonene materials can be favorably utilized. The design strategies and preparation methods of carbonene fibers with regard to the intrinsic scientific and technological processing issues have been briefly reviewed to discuss the opportunities and challenges in bridging the gap between nanoscale carbonene materials and macroscale carbonene fibers. The application of carbonene fibers as lightweight, high-performance fibers, functional fibers, and intelligent fibers has been presented, demonstrating that carbonene fibers are the most promising candidates for next-generation fiber materials due to their unparalleled properties and tremendous applications in extensive advanced engineering fields.

Although spectacular achievements have been accomplished, the development of carbonene fibers faces great challenges. One scientific problem is that the properties of carbonene fibers are still far from those expected. This research status calls for building of a systematic theoretic framework and developing practical processing technologies. Establishing the structure–processing–property relationship will provide guidance for the preparation of high-performance carbonene fibers with designed structures. It is noteworthy that mechanical measurements of carbonene fibers in the laboratory is usually not in conformity with the standards, resulting in a controversial estimation of their properties, since the results largely depend on the testing geometries and methods. Therefore, relevant standards need to be established to objectively evaluate their performances. Another practical problem remains that the vast majority of carbonene fibers are prepared on a laboratory scale. Challenging issues would occur from laboratory to industry, such as the widespread availability of carbonene materials with controlled structures in

large volumes at a relatively low cost and continuous manufacturing techniques for the preparation of high-performance carbonene fibers in large and high quantities. Thus, the combination of fundamental research and industry production is essential. By grasping the law of performance transfer and design principles of advanced carbonene fibers, along with developing the corresponding industrial manufacturing technologies, one can look forward to the ultimate success of carbonene fibers in the near future.

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The manuscript was written through contributions of all authors.

Notes

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