

Building a Bridge for Carbon Nanotubes from Nanoscale Structure to Macroscopic Application

Liu Qian, Ying Xie, Mingzhi Zou, and Jin Zhang*



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ABSTRACT: Through 30 years of research, researchers have gained a deep understanding of the synthesis, characteristics, and applications of carbon nanotubes (CNTs). However, up to now, there are still few industries using CNT as the leading material. The difficulty of CNTs to be applied in industry is the gap between the properties of CNT-based aggregates and those of a single carbon nanotube. Therefore, how to maintain the intrinsic properties of CNTs when they are assembled into aggregates is of great significance. Herein, we summarize and analyze the research status of CNT materials applied in different fields from proven techniques to potential industries, including energy storage, electronics, mechanical and other applications. For each application, the intrinsic properties of CNTs and the real performances of their aggregates are compared to figure out the key problems in CNT synthesis. Finally, we give an outlook for building a bridge for CNTs from nanoscale structure to macroscopic application, giving inspiration to researchers making efforts toward the real application of carbon nanotubes.

1. INTRODUCTION

A carbon nanotube (CNT), a one-dimensional carbon allotrope, is known as a magic material that possesses unique properties and wide application prospects. The structure of a CNT can be regarded as rolling up a single- or multiple-layer graphene nanosheet along a specific vector into a seamless tube,¹ obtaining a single-walled (SWNT) or a multiwalled CNT (MWNT), respectively. The structure, named as chirality, of the CNT can be uniquely determined by the rolling vector. Because of the uncertainty of the rolling vectors, there are great numbers of different SWNTs labeled by their chirality parameters, (n, m) values, which also determine the various electronic and phonon structures of SWNTs. Especially, they can be either semiconducting or metallic according to their chirality. CNTs exhibit outstanding electrical, optical, mechanical, and thermal properties² due to their special structures and nanoscale sizes, thus showing potentials in being applied in the fields of electronics, optoelectronics, energy, military, environment, biology, and so on.^{3,4} Other advantages such as good stability, large specific surface area, and chemical modifiability, also make them compatible with more application scenarios.⁵

After 30 years of study, researchers have made great achievements in the synthesis and applications of CNTs. In terms of structure-controlled synthesis, SWNTs with a specific electrical property and even a specific chirality have been obtained.^{6–8} With regard to mass production, arc-discharge,⁹ floating catalyst,¹⁰ and fluid-bed chemical vapor deposition (CVD)¹¹ methods have been developed to improve the CNT yields. Therefore, CNT powders for energy storage, CNT arrays or films for electronics, and CNT fibers for reinforced composites have all been successfully demonstrated in laboratory research. Nevertheless, except for conducting additives, there are still few industries on the market that

make full use of CNTs. Synthesis determines the future, and the imperfection of material synthesis is the main challenge that the CNT industry faces now. It is the property gap between nanoscale CNT and CNT aggregates that hinders the real application of CNTs.

Metallic CNTs possess a conductivity as high as 10^7 S m^{-1} ;¹² however, when they are used as conducting additives, the architecture of the CNTs in powders greatly affects the performances of batteries due to different conducting efficiencies. An individual semiconducting CNT exhibited superior performance of ultra-high carrier mobility¹³ and low subthreshold swing (~ 70 mV dec^{-1}) even in a 5 nm gate field-effect transistor (FET),¹⁴ exceeding the performance of silicon-based electronics on the same scale. However, CNTs still cannot replace silicon in electronics, in which the purity and mass production of CNT arrays and films are crucial limitations. In addition, the intrinsic tensile strength of an SWNT is predicted to be higher than 100 GPa,¹⁵ but the strongest films that are composed of CNTs showed an average strength of only 9.6 GPa.¹⁶ Realizing good load transfer between tubes when they are assembled into films and fibers is a big challenge. To sum up, the huge gap between the performances of CNT aggregates and nanoscale CNTs mainly originates from the imperfect synthesis and assembly of materials, which lead to the unsatisfactory transfer of the CNT structure and properties. Therefore, how to build a

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bridge for CNTs from nanoscale structure to macroscopic application is a significant issue, as shown in Figure 1.

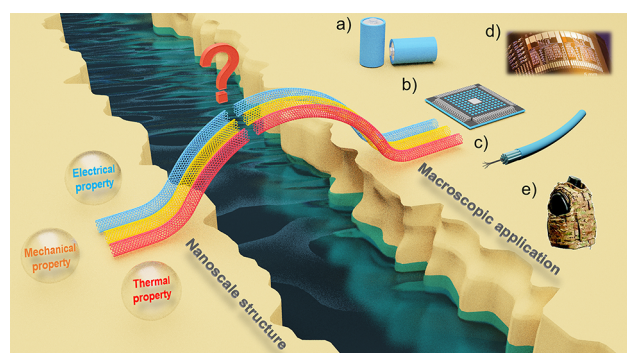


Figure 1. Building a bridge for CNTs from nanoscale structure to macroscopic application is a significant issue. (a) Energy storage. (b) Electronics. (c) Fibers. (d) Flexible electronics. Reproduced with permission from ref 17. Copyright 2008 Springer Nature. (e) Armoring. Reproduced with permission from ref 18. Copyright 2016 Springer Nature.

In this Perspective, we review the properties and performances of CNTs in different application fields, from the relatively mature industry of energy storage to the most focused field of electronics and finally to other promising fields such as fibers. The key problems in CNT synthesis and assembly for different applications are elaborated respectively, and rational strategies to build a bridge from intrinsic properties to aggregation performances and to the real application of CNTs are pointed out. Through careful optimization of synthesis and processing techniques, CNTs

will finally realize their killer application and industrialization in the future.

2. CNTS FOR ENERGY STORAGE

To meet the need of carbon neutrality policy, tremendous efforts have been made to reduce energy-related CO₂ emissions. The huge market demand has led to the rapid development of electric vehicles, which highlights the great importance of developing energy storage systems with high power density, high energy efficiency, and faster charging rate.¹⁹ Lithium batteries and supercapacitors are the most promising power sources, in which the electrodes determine the final performance. Therefore, using advanced electrode materials and designing electrode structures are of great significance. CNTs have excellent electrical conductivity, which favors a decrease in the resistance of energy storage systems. Their good chemical stability helps them to endure high current and voltage. Most importantly, the mass production of CNTs has been proved to be possible. Therefore, CNT is regarded to be one of the star materials for next-generation electrodes.

2.1. Role of CNTs in Energy Storage Systems.

Although CNTs have excellent properties, using them as electroactive materials such as serving as an anode is not an advisable strategy. In comparison with graphite, the most widely used carbon material in an anode, CNTs possess a larger specific area and smaller scale. The capacities of CNT electrodes are 1–2 times that of graphite electrodes according to the report.²⁰ However, the lack of a stable charging voltage plateau and low Coulombic efficiency due to the irreversible insertion of Li⁺ ions become two insurmountable stumbling blocks of CNT anode commercialization.^{21,22} Playing the role of a regulator seems more appropriate for CNTs, which means

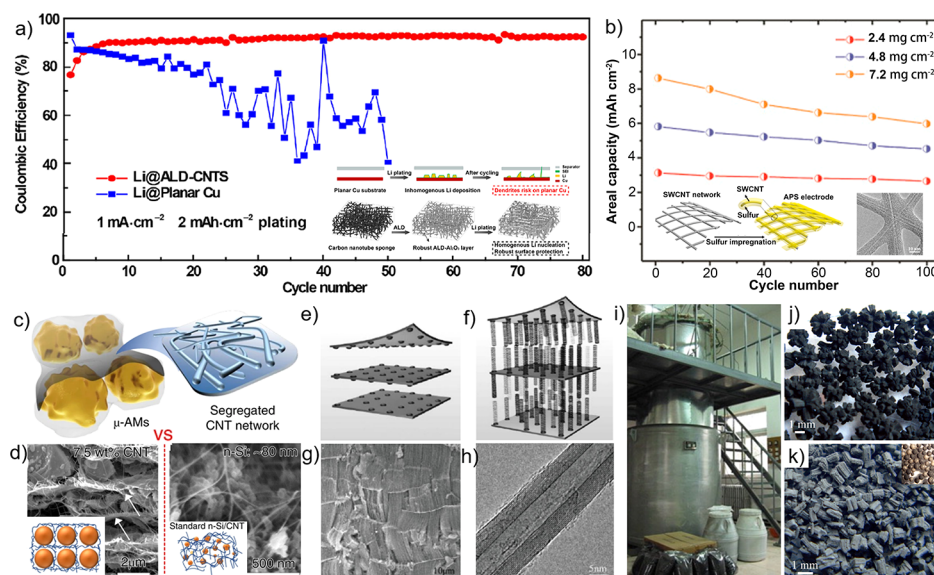


Figure 2. Application of CNTs in energy storage systems and CNT mass production processes. (a) Preparation and electrochemical performance of a 3D CNT sponge@metal lithium anode. Reproduced with permission from ref 24. Copyright 2017 Springer Nature. (b) Preparation and electrochemical performance of Li-S batteries with a SWCNT@S cathode. Reproduced with permission from ref 31. Copyright 2017 Elsevier. (c, d) Fabrication of hierarchical CNT@Si composite electrodes. Reproduced with permission from ref 32. Copyright 2019 Springer Nature. (e–h) Formation process and characterization of hybrid composites by intercalating VACNT arrays into layered inorganic compounds. Reproduced with permission from ref 33. Copyright 2009 John Wiley and Sons. (i) Continuous mass production of CNTs. Reproduced with permission from ref 34. Copyright 2008 Elsevier. (j, k) Separating large-scale CNTs from ceramic sphere substrates through CO₂ oxidation and mechanical vibration. Reproduced with permission from ref 35. Copyright 2010 Elsevier.

they do not react during the electrochemical process but just help to improve the performance of other electroactive materials. To be specific, CNTs can alleviate the growth of lithium dendrites, buffer the volume expansion of an electrode, provide immobilization sites, and serve as ion- and electron-transport facilitators.²³

Safety is one of the most concerning problems for lithium batteries. Numerous explosion and burning accidents have happened because of a short circuit caused by the formation of lithium dendrites. To overcome this issue, Zhang et al. designed a 3D carbon nanotube sponge deposited with lithium metal to serve as an anode. Figure 2a illustrates the preparation process of the anode. CNT agglomerates with a large specific surface area (300–400 m² g⁻¹) supply numerous electrochemically active sites for lithium metal deposition and suppress the formation of lithium dendrites. CNT networks also make the charge distribution of the entire electrode more homogeneous and improve the charge transfer rate of the Li/Li⁺ reaction.²⁴ In addition, compositing with other electroactive materials is a common approach to use CNT materials. It is worth noting that the metal impurities in CNT agglomerates such as metal catalysts must be removed by acid treatment or other ways. Otherwise, the impurities will induce the formation of lithium dendrites.²⁵

In addition to the problem of lithium dendrites, during the process of cycling, with the insertion and extraction of Li⁺ ions, the volume of the electrode will change constantly. The change will destroy the structure of the electrode and result in irreversible capacity decay. Sometimes the volume expansion of an electrode will break the solid–electrolyte interphase (SEI) film and consume more Li⁺ ions permanently, which means lower Coulombic efficiency. Due to CNTs' outstanding mechanical properties, the addition of CNT agglomerates can serve as a framework, improving the structure stability of the electrodes and buffering negative effects of the volume fluctuation during cycling, which is beneficial for increasing the cycle life of batteries.^{22,26}

For both lithium–sulfur (Li-S) batteries and supercapacitors, chemically modified CNTs are often used, especially N-doped CNTs. They can supply enough immobilization sites interacting with the polysulfides because of strong polar–polar surface bonding forming between them, inhibiting shuttle effects and improving the cycling stability of Li-S batteries.²⁷ The key issue for obtaining high-performance supercapacitors is improving their capacitances and energy densities.²⁸ Modifying the surface of CNTs through acid or oxidant treatment can easily introduce oxygen-containing functional groups, which can make the surface of CNTs more hydrophilic and accelerate the process of ions in the electrolyte migrating to the surface of electrode–electrolyte. The modified materials show a pseudo-Faradaic capacitance in the range of 4–135 F g⁻¹.²⁹ Li et al. designed supercapacitor electrode materials with hierarchically structured pores, which were synthesized through carbonizing the composites of ZIF-8 and MWNTs after a treatment with KMnO₄ and HNO₃. At a scan rate of 5 mV s⁻¹, the highest specific capacitance of this supercapacitor is 293.4 F g⁻¹. After 10000 cyclic voltammetry cycles, only 4.2% of the capacitance is lost.³⁰

2.2. CNT Materials for Conducting Additives. For many electroactive materials with low electronic conductivities, such as LiFePO₄ and sulfur, conducting additives are indispensable due to the requirement of rapid charge transfer. Traditional conducting additives such as carbon black and

graphite powder are both 0 dimensional particles, which follow a “point-to-point” conducting mode. In that case, it is necessary to add enough conducting additives to ensure that particles can form effective charge transport paths. In contrast, CNTs follow the “line-to-point” conducting mode, which means a small amount of CNTs can achieve the same effect of carbon black because they can provide long-range transport for electrons and can be incorporated better with active electrode materials.²³ As shown in Figure 2b, Fang et al. used SWNT networks to impregnate with sulfur. Sulfur is uniformly coated on the SWNT networks. The SWNT networks provide abundant routes for Li⁺ ions and electrons to migrate and improve the sulfur utilization. They obtained a high areal capacity of 8.63 mA h cm⁻², which is much higher than that of normal lithium-ion batteries (4 mA h cm⁻²).³¹ In addition, Landi et al. dispersed 1 wt % SWNTs in a LiNi_{0.8}Co_{0.2}O₂ cathode and achieved a high current density of 16.4 mA h cm⁻². The effect surpassed the plan using 4 wt % carbon black as the conducting additive.³⁶ Serving as conducting additives is the most important application of CNTs.

In electrodes, conducting additives do not contribute to the practical energy density directly, so it is crucial to design a specific structure to increase the efficiency of per unit mass conducting additives and decrease the mass ratio of conducting additives in the electrode. According to the experimental results of Park et al., it turns out that conducting additives with a hierarchical structure have much higher efficiency in comparison to those with a disordered structure. They developed a general strategy to construct a hierarchical composite where the active material particles were enveloped by segregated CNT network membranes, as shown in Figure 2c,d. The strategy can be applied to both cathodes and anodes and is compatible with any novel electrode materials. The conductivity of the electrode using that strategy is as much as 1 × 10⁴ S m⁻¹, and the charge-transfer resistance is very low. The resistance of the electrodes with >0.25 wt % CNTs of the segregated network structure is superior to that of traditional electrodes with 20% carbon black/poly(acrylic acid).³² Obviously, we can make the conclusion that ordered and hierarchical structures can decrease the mass ratio of the conducting additives without decreasing the conductivity. Therefore, synthesizing CNT agglomerates with hierarchical structures directly has a very important value. Zhang et al. proposed an approach of intercalating vertically aligned carbon nanotubes (VACNTs) into layered compounds and constructing a layered hybrid nanocomposite (Figure 2e–h). The aligned CNTs were grown from single lamellar particles and avoided the collisions during the growth process; thus, the composites have distinct structures. In addition, these composites have been successfully scaled up in fluidized-bed reactors, realizing the mass production of CNT-based conducting additives.³³ On the basis of this work, Tian et al. designed a magnetically assisted fluidized bed CVD method for the mass production of long CNT arrays. The uniform magnetic field effectively improved the fluidization behavior of catalyst particles. Specially, the magnetic field reduced the bed pressure drop and increased the bed expansion, which helped to accomplish the durative and steady growth of CNT bundles. The yield (9.1 g of CNT/g of catalyst) and length of CNTs (up to ~100 nm) were both improved significantly.³⁷ The aligned CNTs have become a new generation of conducting additives for batteries.

However, the continuous mass production of high-purity CNTs is still a big challenge. Although many research projects have been conducted to explore the basic principle in CNT growth, they were actually carried out in small furnaces and under ideal conditions. There is a huge gap between laboratory experiments and industrial production. The heat/mass transfer, the mechanism of CNT agglomeration, and CNT growth behavior under strong stress all need to be taken into consideration. Many engineering problems need to be solved.³⁸ It is worth noting, however, that mass production of CNTs has made some progress. On the basis of an understanding of the catalytic process, the kinetics of CNT growth, transport phenomena, and so on, the production of CNTs on the ton scale has been achieved through fluidized-bed reactors, as shown in Figure 2i.³⁴ Figure 2j,k illustrates the process of releasing CNTs grown on ceramic spheres by CO₂ oxidation. CO₂ is a proper oxidant that can weaken the interaction between CNTs and the substrates. Meanwhile, CO₂ can also selectively etch away amorphous carbon coated on the CNTs and improve the purity of CNTs. After CO₂ oxidation, it is easy to separate the CNTs and the substrates by a mechanical force.³⁵ A great amount of CNTs can be synthesized through these industrial production processes, which lays a solid foundation for the mass application of carbon nanotubes.

2.3. From a Single CNT to the CNT Industry. If we want to solve the problems that CNTs face at present in the field of energy storage thoroughly, we must have deep insights into CNTs and try to build a bridge connecting materials at different scales, as shown in Figure 3. The first scale is the

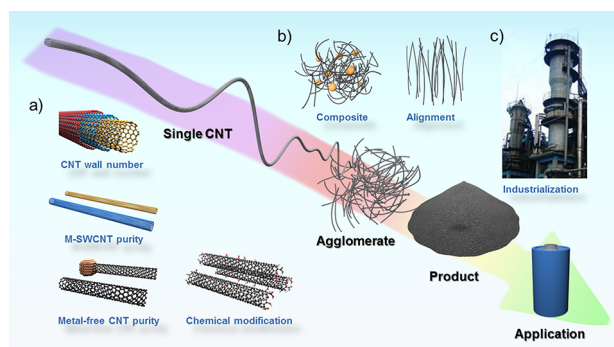


Figure 3. Key points for CNT synthesis and assembly in energy storage. (a) Single CNT. (b) CNT agglomerate. (c) CNT industry. Reproduced with permission from ref 38. Copyright 2013 John Wiley and Sons.

formation of a single CNT on an atomic scale, including the growth mechanism of CNTs. Efforts should be made to control the wall numbers, diameters, and metallic-tube purities of the CNTs through catalyst design and other strategies. In addition, the removal of metal catalysts to obtain metal-free CNTs and the modification of CNTs to introduce functional groups by chemical treatments are both on this scale. The second scale is the structure of CNT agglomerates at a mesoscopic scale. The strong interactions between CNTs influence the agglomeration behavior of CNTs and finally determine the structure of agglomerates. The alignment of CNTs is also affected by the agglomeration behavior, which is the bridge between atomic structure and macroscopic powder products. The last scale is the industrial production and application of CNTs on a macroscopic scale. Macroscopic flow

and heat/mass transfer will influence the production of CNTs. The safety and health problems of CNT products should also be investigated.

The outstanding properties of CNTs indicate great prospects of creating green and safe energy storage systems with high energy density, high power density, and long cycle life. Large numbers of industrial projects on CNT mass production have been carried out in the past 30 years, and the CNT production can reach thousands of tons annually.³⁸ In the future, there must be numerous significant applications in the field of energy storage to fill the gap between the laboratory and life.

3. CNTS FOR ELECTRONICS

Since their discovery,³⁹ SWNTs have long been considered as one of the most promising materials for future electronics due to their outstanding electrical properties.⁴⁰ SWNTs can be metallic or semiconducting according to their chirality, which determines a mixture of about 33.3% metallic and 66.7% semiconducting tubes in ordinary SWNT aggregates. Among them, semiconducting SWNTs (s-SWNTs) are highly desirable for constructing FET-based electronics.

S-SWNTs were estimated to have an intrinsic mobility of as high as $10^5 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at room temperature,⁴¹ which benefits from their perfect one-dimensional structures. The ultralong mean free path of carriers enables a quasi-ballistic transport in SWNTs, which means nearly no carrier scattering or energy dissipation takes place in the body of the tube.^{13,42–44} In addition, because of the high thermal conductivity and good stability of SWNTs, they can carry an ultrahigh current density,^{43,45} ensuring the high performance and excellent stability of CNT-based devices. When SWNTs are used as channel materials in FETs, their ultrathin body (atomic-layer thick) guarantees a superior electrostatic coupling between the gate and the channel, which allows for scaling of the channel length on the basis of an effective gate control, avoiding the short-channel effect.^{42,43} Furthermore, SWNTs possess good compatibility with dielectric materials due to the absence of dangling bonds on their surfaces, which also contributes to better performances of the devices.⁴⁶

On the basis of the above advantages, studies on CNT-based electronics continue to make breakthroughs, including FETs, logic circuits, radio frequency (RF) devices, thin-film transistors, optoelectronics, sensors, and so on.⁴⁰ CNT horizontal arrays and films are both important aggregates for electronic applications. However, to emphasize the potential killer applications of CNTs in next-generation integrated circuits, here we mainly focus on the horizontal-array-based devices, in which CNTs play an irreplaceable role.

3.1. CNT-Based Digital and RF Electronics. Digital and RF electronics are two major applications of CNTs, which take full advantage of the excellent electrical properties of s-SWNTs. Since the first room-temperature FET fabricated from a single SWNT was reported in 1998, researchers have made great efforts to continuously improve the performances and decrease the sizes of single-tube devices. Dai et al. used the high-dielectric-constant material zirconium oxide (ZrO₂) as the gate dielectrics for individual s-SWNT FETs, which resulted in subthreshold swings (SS) of $\sim 70 \text{ mV decade}^{-1}$ for p-type transistors, approaching the room-temperature theoretical limit.⁴⁷ They further realized the ballistic transport in a p-type SWNT transistor by greatly reducing the contact barriers using palladium (Pd) as the contacting electrodes.¹³

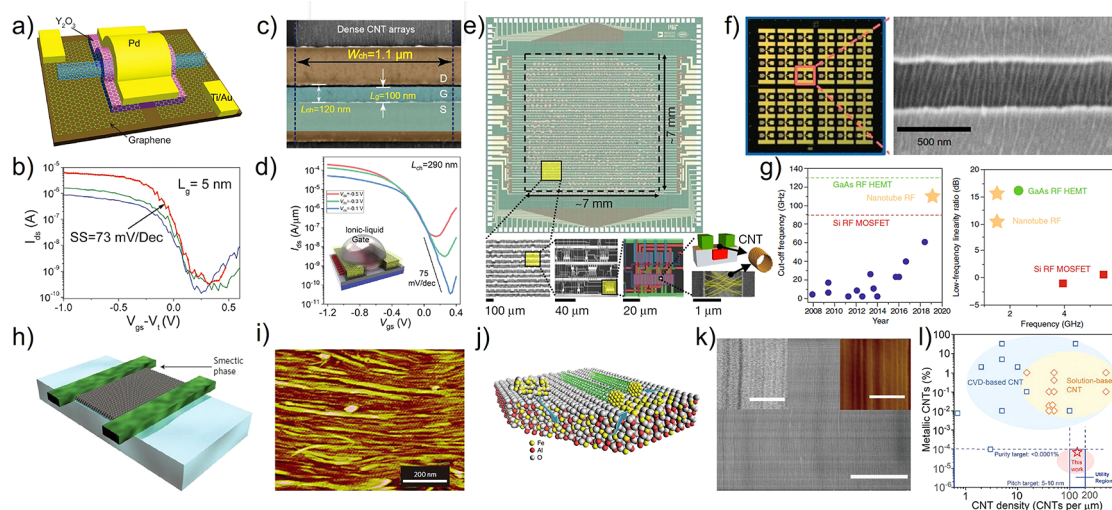


Figure 4. CNT-based digital and RF electronics and high-density CNT horizontal arrays. (a, b) Schematic diagram and transfer curves of the 5 nm gate length CNT FETs. Reproduced with permission from ref 14. Copyright 2017 the American Association for the Advancement of Science. (c, d) SEM image with a detailed channel region and top-view structure and the transfer characteristics of a typical CNT array-based FET. Reproduced with permission from ref 54. Copyright 2020 the American Association for the Advancement of Science. (e) Image of a fabricated RV16X-NANO chip. Scanning electron microscopy images with increasing magnification are shown below. Reproduced with permission from ref 60. Copyright 2019 Springer Nature. (f) Optical micrograph of an array of CNT FETs for RF electronics and SEM images of a channel. Reproduced with permission from ref 63. Copyright 2019 Springer Nature. (g) The cutoff frequency (left) and the linearity figure of merit (right) of the CNT RF transistors. Reproduced with permission from ref 64. Copyright 2019 Springer Nature. (h, i) The LS approach and AFM images of the obtained CNT arrays. Reproduced with permission from ref 66. Copyright 2013 Springer Nature. (j, k) Schematic illustration and the results of growing high-density horizontally aligned single-walled carbon nanotube arrays using Trojan catalysts. Reproduced with permission from ref 67. Copyright 2015 Springer Nature. (l) Semiconducting purity versus density of CNT arrays based on the reported methods. Reproduced with permission from ref 54. Copyright 2020 the American Association for the Advancement of Science.

Peng et al. promoted the realization of a ballistic n-type SWNT FET by contacting s-SWNTs using scandium (Sc).^{44,48} Better n-type FET performances with an SS value of 73 mV/decade and an electron mobility of $5100 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ were obtained using yttrium (Y) as the contacting metal.⁴⁹ On the other hand, the channel length of SWNT transistors scaled from hundreds of nanometers to 15 nm,⁴² sub 10 nm,^{50–52} and finally 5 nm,¹⁴ which might be the ultimate scaled limit on consideration of the performance and power consumption.⁴⁶ The 5 nm gate length transistor (Figure 4a) was fabricated using graphene contacts, which exhibited better performance and less energy consumption in comparison to silicon (Si) complementary metal oxide semiconductor (CMOS) FETs on the same scale (Figure 4b).

Although single-tube transistors have exhibited outstanding performances approaching the theoretical limit, future electronics based on CNTs would require multiple semiconducting tubes per channel to ensure the driving ability in digital circuits. Arnold et al. reported a quasi-ballistic CNT transistor fabricated from CNT arrays of 47 tubes μm^{-1} through floating evaporative self-assembly from sorted semiconducting CNTs, whose on-state current density exceeded those of Si and GaAs FETs.⁵³ Recently, a dimension-limited self-alignment (DLSA) method was developed and used to prepare aligned SWNTs (semiconducting purity >99.9999%) on 4 in. silicon wafers. Top-gated FETs based on the arrays (Figure 4c) exhibited better performances in comparison to commercial Si devices with similar gate lengths (Figure 4d),⁵⁴ promoting the progress in the scalable production of high-performance CNT FETs.

On the basis of the success in FETs, CNT-based logic circuits have also made major breakthroughs.^{55–57} A milestone came when the first CNT computer was developed in 2013,⁵⁸

which was composed of 178 CNT FETs and could run basic operation systems. The aligned CNT arrays used in the computer were grown on quartz substrates, and >99.99% of metallic tubes were removed through electrical breakdown to ensure the performance and logic functionality.⁵⁹ The modern microprocessor RV16X-NANO built from >14000 complementary CNT FETs came out in 2019, which operated standard 32-bit instructions (Figure 4e).⁶⁰ The CNTs used in this microprocessor were deposited from purified CNT dispersions (semiconducting purity of 99.99%) with 40–60 CNTs per linear micrometer. The intrinsic material imperfection was actually overcome by device design techniques.

RF electronics are important systems for wireless communication, which are used for amplifying and switching high-frequency signals. CNT transistors are promising for RF electronics because of the high carrier mobility, high current-carrying ability, and small intrinsic capacitance of SWNTs.^{61,62} Therefore, a high density of parallel SWNTs with a high purity of semiconducting tubes is also necessary. A wafer-scalable fabrication of aligned CNT FETs operating at over 100 GHz frequencies was reported, in which the CNT arrays were fabricated through a floating evaporative self-assembly process (Figure 4f).⁶³ With optimized processing of both materials and devices, the CNT RF transistors can outperform Si RF transistors and approach the GaAs high-electron-mobility transistors (HEMTs) in both cutoff frequency and linearity (Figure 4g).⁶⁴ We are on the way to CNT-based practical electronic systems, and there is still plenty of room because of the imperfect materials.

3.2. From Single CNT to Horizontally Aligned CNT Arrays. It is obvious that the progress in CNT electronics is greatly related to the material preparation techniques, and the

imperfection of materials is a major bottleneck limiting the development of the CNT electronics industry. In the aforementioned cases of successful CNT integrated circuits, in order to achieve excellent performance, researchers purified and aligned carbon nanotubes by electrical breakdown⁵⁸ or solution processing,⁶⁰ which greatly damaged their quality, density, and alignment and introduced contaminations, causing a gap between a CNT array device and a single CNT device. Therefore, how to build a bridge from a single CNT to horizontally aligned CNT arrays applied in electronics remains a problem.

Issues in the past decades attracting the most focus are the density and semiconducting purity of horizontally aligned CNT arrays, because a digital circuit requires FETs built from enough parallel s-SWNTs to realize a high operation speed and logic ability.⁴⁵ Current methods for preparing high-density horizontal arrays mainly include postprocessing and direct growth methods. Postprocessing means reassembling CNTs with a low density or undirected alignment onto the target substrates by transferring or rearranging to obtain high-density arrays. Representative approaches of solution rearrangement include the Langmuir–Blodgett (LB)⁶⁵ and Langmuir–Schaefer (LS)⁶⁶ techniques (Figure 4h), which have been used to achieve a full-coverage SWNT array with a density of up to 500 tubes μm^{-1} from CNT dispersions with 99% semiconductor purity (Figure 4i). The DLSA method mentioned before is also a solution-processing technique.⁵⁴ Other postprocessing methods, such as multiple transfer⁶⁸ and DNA directed techniques,^{69,70} are usually complicated and hard to realize in mass production, which also limit their wide application. The direct growth method refers to the growth of high-density SWNT arrays by chemical vapor deposition (CVD) using the orientation effect (crystal-lattice or atomic-step orientation) of a specific substrate.² Representative strategies for the direct growth method include multicycle growth,⁷¹ multiple deposition of catalyst strips,⁷² *in situ* loading of catalysts,⁷³ etc. The highest densities of 130–160 tubes μm^{-1} were obtained through a Trojan catalysts method, which realized the slow release of iron catalysts from the sapphire substrates (Figure 4j,k).^{67,74} The direct growth methods do not require complex postprocessing, and thus the obtained arrays are clean, are of high quality, and have good alignment.

Similarly, there are mainly two ways to improve the purity of s-SWNTs, post-treatment and direct control during growth. Post-treatment means obtaining a high purity of semiconducting tubes from the mixed carbon nanotubes either on substrates or in solution. The as-grown arrays can be purified by selectively removing the metallic tubes through breakdown,^{75–77} etching,^{78,79} or separation,^{80,81} which obviously decrease the density of arrays. Solution separation is now a mainstream approach to prepare CNT materials for device fabrication due to its high efficiency and good result (>99.99%) for sorting of s-SWNTs. Representative methods include density gradient ultracentrifugation (DGU),⁸² gel chromatography (GC),⁸³ aqueous two-phase extraction (ATPE),⁸⁴ and selective dispersion (SD).^{85,86} Solution separation methods rely on the amplification effect of the differences in CNTs with different electrical or chiral structures using molecules such as polymers and DNA chains and the subsequent separation under strong external fields, thus introducing impurities and damages to CNTs and further affecting the device performances. In addition, the aligned assembly and purification processes are necessary before device

fabrication, which might cause the misalignment and bundling of CNTs, decreasing the efficiency of devices. Therefore, direct control strategies during growth are much preferred in future device applications.

Current methods for selectively growing s-SWNTs directly in CVD mainly include catalyst design, *in situ* etching, and others. Catalysts are significant in the control over CNT structures because they are actually the templates for tube growth. Bimetal catalysts,⁸⁷ solid catalysts,⁸⁸ and oxide catalysts⁸⁹ have been developed to effectively improve the s-SWNT contents in CVD growth. Especially, the solid catalysts can maintain their crystal structures under high temperature, thus thermodynamically controlling the structures of grown SWNTs with specific symmetry and even specific chirality.^{6,90} *In situ* etching methods work by introducing etchants from the growth atmosphere,^{91,92} catalysts,^{93,94} or external sources⁹⁵ during growth, which also lead to a decreased array density. Other methods such as the electrorenucleation (ERN) approach⁹⁶ and rate-selected growth⁹ have been developed recently, which amplified the band-structure differences between s- and m-tubes through an electrical field and ultralong growth, showing high purities of 99.9% and 99.9999% for s-SWNTs, respectively. The two methods reveal the potential for selectively growing ultrahigh-purity semiconducting tubes but also suffer from low density. Figure 4l demonstrates the data of densities and semiconducting purities from some reported methods, indicating the great challenge in obtaining high density, high semiconducting purity, and high quality at the same time.

3.3. Potential CNT Materials for Application in Electronics. On the basis of the research status of CNT electronics and CNT materials used in electronics, we still face some major challenges before their real application. As shown in Figure 5, there are several cracks in the line connecting a

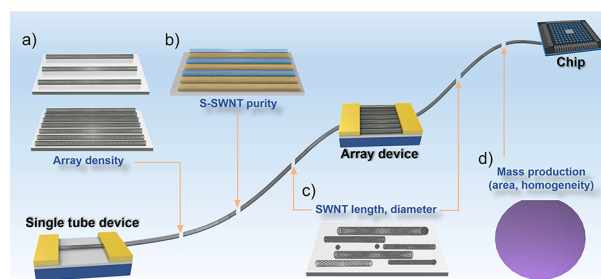


Figure 5. Key points for material synthesis from a single-tube device to a CNT chip. (a) Array density. (b) Purity of s-SWNTs. (c) Lengths and diameters of the SWNTs. (d) Mass production.

single CNT device to a CNT-based chip, representing the obstacles on the way to a CNT electronics industry. As mentioned previously, the two most concerned and studied issues are the density and the semiconducting purity in an SWNT array (Figure 5a,b). It was predicted that a parallel SWNT array with a density of 100–200 tubes μm^{-1} (a small pitch of 5–10 nm) and an s-SWNT purity of 99.9999% is needed for an integrated system to drive the demand current^{45,97} and outperform the conventional Si in digital and RF electronics.⁶¹ Among all the strategies in section 3.2, solution processing is approaching the goal through multiple dispersion and sorting,⁵⁴ but the damage of tube length and the polymer residues leads to a decreased efficiency and poor

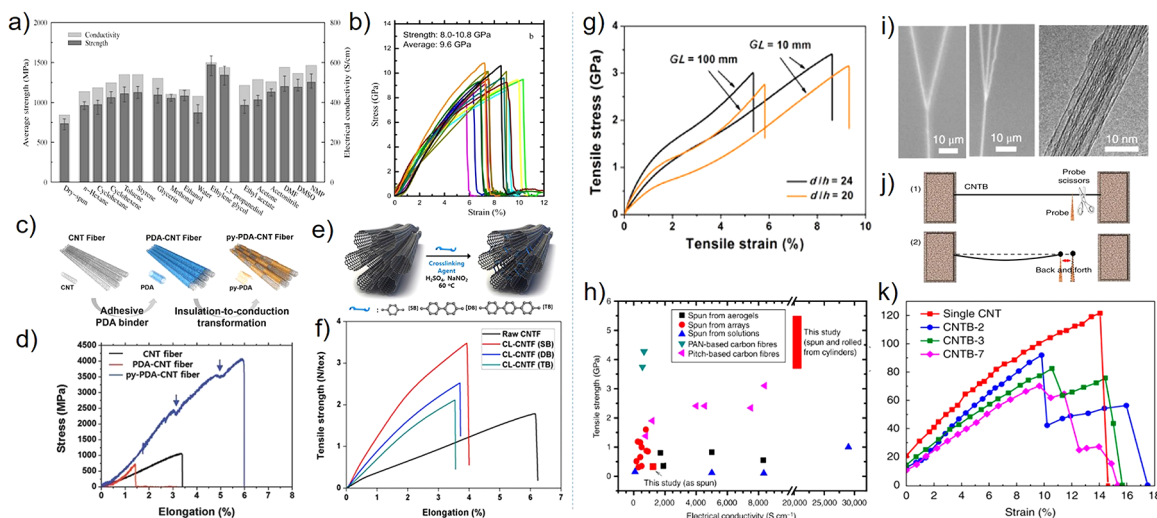


Figure 6. Assembly and performances of CNTFs. (a) Tensile strength and electrical conductivity of CNT fibers after different solvent infiltrations. Reproduced with permission from ref 123. Copyright 2012 Elsevier. (b) Stress vs strain curves showing a high tensile strength of 9.6 GPa for CNTF. Reproduced with permission from ref 16. Copyright 2016 American Chemical Society. (c) Schematic image of compositing the CNTF with pyrolysis-polydopamine (py-PDA). (d) Force–distance curves of the py-PDA-CNTF composite. Reproduced with permission from ref 125. Copyright 2015 John Wiley and Sons. (e) Schematic diagram of cross-linked CNTF. (f) Stress–strain curves of the cross-linked CNTF. Reproduced with permission from ref 127. Copyright 2017 Elsevier. (g) Stress–strain curves of two fibers tested at two gauge lengths (10 and 100 mm), respectively, demonstrating the high tensile strength to be almost retained at a longer gauge length. Reproduced with permission from ref 130. Copyright 2017 Elsevier. (h) CNTF densified by rolling exhibiting both high tensile strength and good electrical conductivity. Reproduced with permission from ref 124. Copyright 2014 Springer Nature. (i–k) CNT bundles (CNTBs) treated with the synchronous tightening and relaxing method: (i) SEM and TEM images of single CNTs and CNTBs; (j) mechanism of the synchronous tightening and relaxing treatment; (k) stress–strain curves for single CNTs and CNTBs with a tensile strength of 80 GPa. Reproduced with permission from ref 131. Copyright 2018 Springer Nature.

contacts between CNTs and electrodes. As a result, an SS value of $190 \text{ mV decade}^{-1}$ was obtained,⁵⁴ much higher than that of the requirements and of the single-tube performance ($\sim 70 \text{ mV decade}^{-1}$).¹⁴ Therefore, the ideal way to prepare SWNT arrays for electronics is direct growth on substrates. Although it is still hard to meet both high density and high s-SWNT purity currently, the combination of catalyst design, substrates, growth atmosphere, and external fields may contribute to the final goal.

In addition, the length and diameter distribution (Figure 5c) of the CNTs in the array should also be taken into consideration in the pursuit of high-performance devices, especially highly integrated circuits, in which device-to-device uniformity is important.⁹⁸ SWNTs with uniform and sufficient lengths in arrays contribute to the best use of all the tubes in the channel and uniform performances in integrated circuits; thus, solution processing for array assembly cannot meet the requirement. The diameters of SWNTs are related to the band structures and current-carrying capability of the tubes,^{99,100} affecting the contacts and the threshold-voltage variations of FETs.⁹⁷ It was reported that SWNTs with relatively large diameters can form better contacts with metals with high work functions,¹⁰¹ and a small range of threshold voltages can have great effects on the leakage current.⁹⁷ Therefore, appropriate diameters of 1.2–2.0 nm and a small variation in diameter distribution are demanded for high performance (on-state currents) in individual FETs and less energy consumption in integrated circuits.⁹⁷ The control of SWNT diameter in CVD growth can be realized by controlling the catalyst sizes and structures, such as monodispersed catalysts,¹⁰² size-separated ferritin-based catalysts,¹⁰³ and solid catalysts,^{88,90} which reveals the importance of catalyst design in SWNT preparation.

A path that must be traveled before the realization of CNT-based chips is the mass production of high-density semi-conducting SWNT arrays (Figure 5d). CNT materials that can be applied in the electronics industry must be wafer-scale, high-density, high-quality, and uniform arrays of s-SWNTs with narrow diameter distributions. The increase in high-density area and the density homogeneity are big challenges in the industrialization of CNT-based chips. Although a 10 cm-scale silicon wafer was successfully covered by high-density and high-semiconducting-purity SWNT arrays through solution separation and assembly,⁵⁴ the solution processing dramatically degraded the qualities of CNTs. Therefore, great efforts should be made in the direct growth of wafer-scale SWNT arrays for electronics. The better uniformity of substrates, better distribution of catalysts, and better homogeneity of growth fields must be considered. A magnesium-assisted catalyst anchoring strategy was developed recently to greatly increase the density uniformity in large-area high-density SWNT arrays,¹⁰⁴ offering a new idea for optimizing the catalyst sizes and distributions on substrates. Many research breakthroughs have brought hope to CNT electronics and show the potential of real application of CNTs in future chips.

4. CNTS FOR MECHANICAL APPLICATIONS

CNTs are predicted to have superb mechanical properties due to their strong carbon–carbon covalent bonds and seamless cylindrical graphitic structure. Theoretically, CNTs are simultaneously endowed with high strength (100–200 GPa), high modulus (1–2 TPa), and high breaking strain ($>15\%$).^{15,105–108} However, these high mechanical performances are not always experimentally available, especially on the macroscale. This is mainly due to the defects in CNTs, which

poses a great challenge to the controlled synthesis. Once CNTs possess perfect structures, for example, the tensile strengths of the superstrong, ultralong CNTs can reach up to 200 GPa.¹⁰⁹ The outstanding mechanical and physical properties of CNT individuals have motivated researchers to develop high-performance macrostructures based upon CNTs, such as CNT fibers (CNTFs).

4.1. CNT Fibers. The CNTF is a typical one-dimensional macroscopic assembly aggregated from nanometer-sized individuals, possessing many outstanding properties, especially mechanical properties. Solution,^{110–112} forest,^{113–116} and aerogel^{117–121} spinning are three main methods to assemble individual CNTs into a fiber. In solution spinning, CNTs are dispersed into certain solutions and then injected into a coagulation bath. In forest spinning, the vertically aligned CNTs grown on substrates are drawn like a thread from a silk cocoon and assembled. In aerogel spinning, individual CNTs are collected directly after synthesis from a furnace chamber through floating catalyst CVD. All three methods have both pros and cons. Highly aligned and densified CNTF could be obtained, but an assembly by short and poorly dispersed CNTs features prominently in the solution spinning. For the forest spinning, the bundles are mostly aligned but the packing density and size control remain challenging. For the aerogel spinning, small and strongly entangled CNTs, the largely variable fiber diameters and embedded byproducts, such as Fe particles and certain organic compounds, could weaken the superb performance of the fibers. Although the aforementioned spinning methods still face challenges to perfectly align and highly pack CNTs, liquid crystal spinning has solved this problem to some extent. The CNTs, due to their rigid-rod-like shape, can form a liquid-crystal phase under certain conditions. Therefore, liquid-crystal spinning is promising in achieving high nanotube orientation for CNTF fabrication.¹²² However, such a spinning method is confronted with challenges such as a high dispersion of CNTs for liquid-crystal phase formation. All in all, a facile method compatible with all requirements for assembling CNTFs with higher packing density and better alignment is still highly desired. Altogether, the waviness, poor alignment, and weak intertube interactions of CNTs still remain the most remarkable restrictions and significantly deteriorate the load transfer efficiency. Thus, the densification of CNTs in front of the fiber formation is a desirable way to solve this problem.

The first effective way is solution densification, where the slow evaporation of highly polar solvents attracts CNTs by a capillary force. Typical solvents used in condensation are given in Figure 6a, and the most effective one is ethylene glycol, resulting in a tensile strength of 1.45 GPa.¹²³ The second method is mechanical densification. The most typical example was given by Wang et al. in 2014, where they designed a pressurized roller to densify the initial fiber, leading to a high tensile strength of 4.34 GPa.¹²⁴ Figure 6b shows that an average strength of 9.6 GPa can be reached by the imposition of mechanical rolling.¹⁶ Thirdly, polymer infiltration also has a better strengthening result. For example, a bismaleimide (BMI)-infiltrated CNTF could exhibit an enhanced strength of 2.38 GPa.¹²³ As shown in Figure 6c,d, when a graphene-like polymer, pyrolysis-polydopamine (py-PDA), which strongly interacts with CNTs by a graphitic interaction, was composited, the py-PDA-CNTF showed a further enhanced tensile strength of 4.04 GPa.¹²⁵ Finally, cross-linking has been proven to be efficient as well. The linking effects can stem from

the direct shell–shell and tube–tube bonding, such as irradiating double-walled carbon nanotube bundles with 200 keV electrons in transmission electron microscopy (TEM) enhanced the tensile strength up to 17 GPa, but scaling up would be difficult for this processing.¹²⁶ Introducing additional linkers between CNTs is more common. For instance, in Figure 6e,f, the tensile strength of CNTF after a 2 h aryl cross-linking reaction was comparable to that of Toray T1100 grade (3.7 N/tex), which is the strongest commercial carbon fiber in the world.¹²⁷ Other linking mechanisms, such as a π – π conjugation with the aromatic structure of the CNT¹²⁸ and covalent bonding through fluorinated CNT free radical chemistry,¹²⁹ both enhanced the tensile strength of CNTF to different extents, showing values of 1.7 and 1.38 GPa, respectively.

4.2. From a Single CNT to High-Performance CNT Fibers. When CNTs are assembled into fibers, the structural homogeneity will have a great effect. The mechanical properties of CNTF can be strongly affected by gauge length. For example, a super high strength of 8.8 GPa has been reported as early as 2007, but it was measured with only 1 mm gauge length. Thus, fabricating CNTFs with long-range structural homogeneity is crucial for their real applications. The last few years have witnessed the enhancement of the homogeneity of CNTF. The tensile strength of aryl cross-linked CNTF (3.7 N/tex) was almost unchanged using samples of 1, 5, and 10 mm gauge lengths.¹²⁷ A high average strength of 4.34 GPa after rolling processing was also obtained with a sample of 10 mm gauge length and did not change much with 20 mm gauge length.¹²⁴ As shown in Figure 6g, after a similar twisting and compressing process, the tensile strength still remained at relatively high values of 2.5–3.0 GPa with 100 mm gauge length.¹³⁰ In order to enhance the homogeneity of CNTFs, initial CNTs with coherent length are needed. When they are bundled together, a high degree of axial alignment and high packing with few voids are required as well.

Another concern in CNTF synthesis is enhancing the mechanical properties to the maximum without sacrificing electrical conductivity. Among the four methods of densifying CNTs, although the polymer compositing or cross-linking compacts the voids between individual CNTs which may improve the conductivity to some extent, the insulating polymer networks or linkers tend to block the electron-hopping channels between CNTs and deteriorate the conductivity of CNTF. For instance, an aryl cross-linked CNTF¹²⁷ and a cured BMI-CNTF¹²³ composite both claimed an improved conductivities of 1.4×10^3 and $\sim 0.88 \times 10^3$ S cm^{-1} , respectively, far below that of individual CNT itself. To further eliminate the effect of an insulating polymer, an electrically conductive polymer (py-PDA) was applied to enhance the conductivity to 5×10^3 S cm^{-1} .¹²⁵ Nevertheless, no matter how carefully the polymer or linker was designed, their use inevitably compromised the purity and conductivity of the CNTF. A better result was achieved by rolling-densified CNTs with both high tensile strength (4.34 GPa) and good conductivity (2×10^4 S cm^{-1}), as shown in Figure 6h.¹²⁴ On combination of the discussion of the homogeneity earlier with that of the conductivity here, it seems mechanical densification is a better choice. Similar to a mechanical processing mechanism, Fei et al. crafted CNT bundles with a length of centimeters and a tensile strength of over 80 GPa (corresponding to an engineering strength of 43 GPa) by synchronously tightening and relaxing the initial CNT bundles several times

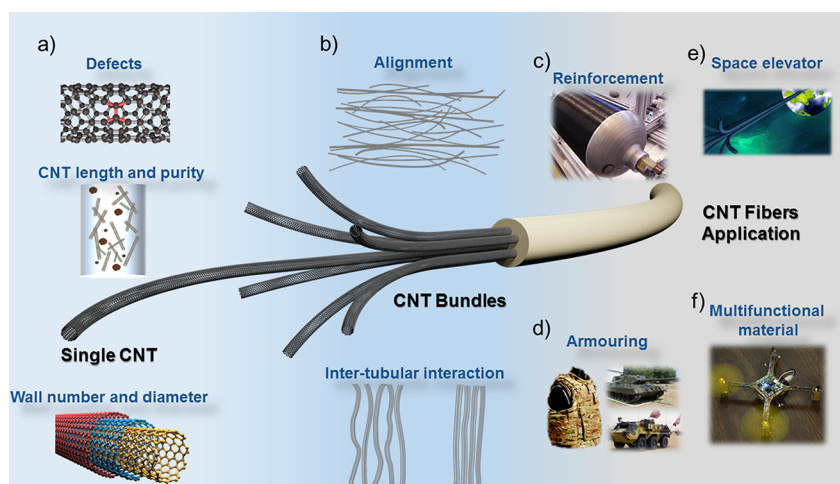


Figure 7. Key points for the synthesis and assembly from a single CNT to CNTFs and potential applications of CNTFs. (a) The properties of single CNTs should be considered for constructing an ideal CNTF. (b) Problems in CNT bundles, which may block the transfer channel of the superb performance from single a CNT to a CNTF. (c–f) Potential applications of CNTFs. (c) Tension-dominated areas. Reproduced with permission from ref 132. Copyright 2016 Elsevier. (d) Armouring, Reproduced with permission from ref 18. Copyright 2016 Springer Nature. Reproduced with permission from ref 134. Copyright 2010 Elsevier. (e) Space elevator. (f) Multifunctional material. Reproduced with permission from ref 135. Copyright 2016 Elsevier.

to release the different initial strains (Figure 6i–k). They claimed that nonuniform initial strains were the principal reason for a remarkable decrease in the tensile strength for CNTF. Unfortunately, they failed to give the electrical properties of their super strong fiber.¹³¹

By briefly reviewing the CNTF fabricating technique, we modularize the macrostructure by breaking it into three parts: from individual CNTs to the bundles and finally to the fibers. Enhancing the tensile strength of CNTFs primarily requires a consistent production of long, perfectly structured, and single- or few-walled CNTs with high purity (Figure 7a). When they are assembled, as shown in Figure 7b, a good alignment and elimination of voids between CNTs are required. Therefore, certain methods are needed to enhance intertubular interactions, thus improving the load-bearing capacity, as has been discussed in detail in section 4.1. Once the intertubular interactions within CNTF can be maximized, the theoretically predicted strength of an individual CNT may be reachable in the macroscopic fiber, which will greatly enlarge the applications of CNTF in the near future.

4.3. Potential Applications of CNT Fibers. CNTFs with multiple properties widely surpassing those of traditional materials has stimulated the imagination of applications in many advanced fields, such as aviation, automobiles, reinforcement for tension-dominated applications, and defense industries, such as armor for both military and civilian areas. CNTFs with high tensile strength can be exploited as reinforcements for lightweight, high-strength, multifunctional composites in tension-dominated applications such as composite overwrapped pressure vessels (COPVs). An Epon828/CNT yarn composite overwrapped pressure vessel, shown in Figure 7c, could exhibit a 209% increase in hoop tensile properties with only 10.8 wt % of CNTF/Epon 828 wrapping over Al rings. With the improved strength-to-weight ratio, the reinforced COPVs could yield significant weight savings that minimize the mass of spacecraft and launch vehicles.¹³² In addition, the interest in exploring outer space or ever-increasing threats such as terrorism faced by mankind currently have spurred the development of much more reliable

armor protection for airplanes, tanks, combat vehicles, and the human body (Figure 7d). The ballistic resistance requires lightweight materials with extremely high tensile strength and modulus. Previous research has demonstrated that reinforcing the matrix with a very small amount of CNTFs resulted in much more resistance to penetration of a projectile by effectively absorbing the kinetic energy locally.¹³³ Furthermore, on the basis of their superb structural and mechanical properties, CNTs have been envisioned as being promising for tethers of space elevators. Providing access to earth orbit via a 144000 km long cable requires it to sustain a stress of 63 GPa with a density of 1.3 g/cm³. Only CNTs are possible of possessing such extraordinarily high strength (Figure 7e).^{136,137} In addition to maximizing certain properties of CNTFs in advanced applications, realizing multifunctionalization is highly desirable as well. For instance, in Figure 7f, a quadcopter frame manufactured from continuous CNT yarn reinforced poly(ether imide) (Ultem) by additive manufacturing (AM) demonstrates the potential of taking advantage of the diverse properties of CNTFs to impart multiple functionalities, such as mechanical reinforcement, electrical conductivity, resistive heating, strain sensing, etc., to tailored components.¹³⁵ In a word, once the anticipated mass production of multifunctional materials imparted by CNTF addition is realized, the domination of CNTF-based materials in both military and civilian areas would be foreseeable in the near future.

5. OTHER APPLICATIONS OF CNTS

Endowed with numerous superior properties, CNT puts itself in the limelight of being applied in many other fields, such as electromagnetic shielding, thermal management etc. Most traditional composites for electromagnetic shielding have high density and suffer from poor temperature and corrosion resistance. These drawbacks can be alleviated by CNT addition due to its tunable structure, light mass, and excellent electrical conductivity. When it is highly dispersed in a matrix, the added CNT enables great attenuation of microwaves at a much lower filler fraction and a thinner thickness. In addition, judiciously

designed CNT aggregates could bring about additional properties as well. For instance, a large-sized CNT sponge welded by amorphous carbon was designed to construct ultrablack materials with omni-antireflection (low reflectivity of 0.96–0.41% in a wide wavelength range of 400–1500 nm). The porous structure (99%) made it ultralight (2.1 mg cm^{-3}), and amorphous carbon joined between CNT individuals simultaneously endowed the sponge with outstanding mechanical durability and elastic flexibility, which is beneficial for military applications.¹³⁸

Another potential application of CNTs is being used as a thermal interfacial material (TIM), which is applied between two interfaces to realize continuous and good heat dissipation. A thermal interfacial material is quite important for the sustainable operation of electronic systems such as integrated circuits.^{139,140} The use of CNTs in TIMs relies on their good thermal and mechanical properties. CNTs have high thermal conductivity and can be developed onto different substrates. Other advantages such as good chemical stability, low coefficient of thermal expansion, and low transverse elastic modulus all contribute to their excellent heat dissipation performance.^{141,142} Vertically aligned CNTs are the main aggregates used as TIMs because they are CNT forests with highly packed density. Apart from pure CNT TIMs,¹⁴³ many efforts have been put into the moderation and composition of CNTs to improve the heat transmission. For example, when three-dimensional networks are built for higher thermal capacity,¹⁴⁴ CNT/polymer composites have a comprehensive performance.¹³⁹ It is believed that through the design of materials, the CNT TIMs will be useful in many areas of our modern life.

6. SUMMARY AND PROSPECTS

In summary, research on CNTs have permeated into many different fields related to daily life and the military. In the field of energy storage, the production and application of CNTs is a relatively mature industry. The optimization of CNT assembly and technical problems in mass production considering cost and performances may be the most important issues. However, the role of CNTs in energy storage is actually as an additive, which cannot take full advantage of their outstanding properties. In the field of electronics, SWNTs exhibit outstanding performances in laboratory research, showing great promise for future industry. The controlled synthesis of demanded SWNT arrays and large-scale production are urgent problems to be solved. CNT fibers, electromagnetic shielding, and thermal interfacial materials are other potential fields where CNTs will play important roles. The suitable assembly and moderation of CNT aggregations will promote their progress in real applications. In a word, how to build a bridge from a nanoscale structure to a macroscopic application is a common challenge that CNT materials face now. On the basis of the research status above, we put forward an “IDEA” strategy to pursue better property transfer when preparing CNT aggregates for applications. As shown in Figure 8, the puzzle indicates that each piece of “IDEA” is an integral part of composing a perfect aggregate.

“I” means interaction, which refers to the interaction between monomers when CNTs are assembled into aggregates or into composites with different materials, including interaction forces, possible charge transfer, etc. The interaction between monomers has a great effect on the final properties of the aggregates because it is related to the property transfer and

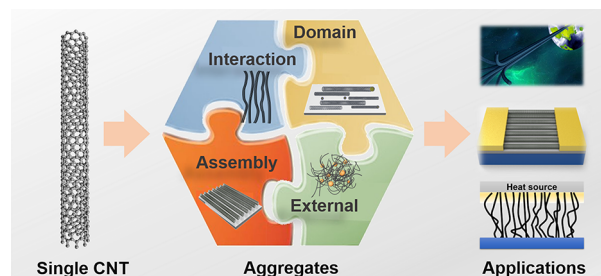


Figure 8. An “IDEA” strategy for property transfer from a single CNT to CNT aggregates and finally to applications.

reveals the short plank of an aggregate. For example, in CNTFs, better intertubular interaction is an important point to improve the tensile strength of the fibers. Mechanical densification and cross-linking are effective ways to obtain stronger fibers through better intertubular interactions. The transfer of electrical properties can also be optimized through better interactions, such as carbon-welded SWNTs for high-performance transparent conducting films.¹⁴⁵

“D” means domain, which refers to the basic function unit in an aggregate, including the domain size, morphology, quality, etc. The domain in an aggregate undertakes the basic function of the material; thus, an appropriate size and morphology, high quality, and necessary moderation are all important factors affecting the final performances of the aggregates. For example, when high-performance FETs are fabricated from SWNT horizontal arrays, the electrical properties, lengths, diameters, and quality of the SWNTs should all be taken into consideration. In CNTFs, increasing the average lengths of the CNTs is also an important way to improve the structural homogeneity and mechanical properties of the fibers.

“E” means “external”, which refers to the necessary introduction of external fields or materials into CNTs. Although CNTs have outstanding properties in many aspects, the introduction of externals sometimes can induce a synergistic effect and contribute to better performances of the aggregates. Studies on compositing CNTs with other materials like graphene and polymers are often reported,^{146,147} which take advantages of both the materials and endow the composites with higher performances and additive functions.

“A” means “assembly”, which refers to the assembly structures of the aggregations, including the dimension, alignment, architectures, etc. The assembly structures of CNTs are the first element to be determined in the construction of aggregates for certain applications. On the basis of the application scenarios of the materials, suitable or unique structures of CNT aggregations should be considered, such as horizontally aligned arrays for FETs and vertical arrays for heat dissipation. In the CNT conducting additive industry, the assembly of vertically aligned CNTs between layered compounds is a great way forward for new-generation CNT conducting additives.³³

Therefore, we should optimize the interactions between tubes, the domains of the aggregates, the necessary introduction of externals, and the assembly structures of CNTs to realize better property transfer from nanoscale tubes to macroscopic aggregates, which is the so-called “IDEA” strategy. The “IDEA” strategy offers a direction when CNT aggregates are constructed for specific applications or the current CNT materials are optimized for future industry. Through building this bridge for CNTs from a nanoscale

structure to macroscopic applications, there may be a day that presents a brand-new era of carbon.

AUTHOR INFORMATION

Corresponding Author

Jin Zhang – Center for Nanochemistry, Beijing Science and Engineering Center for Nanocarbons, Beijing National Laboratory for Molecular Sciences, College of Chemistry and Molecular Engineering, Peking University, Beijing 100871, People's Republic of China; orcid.org/0000-0003-3731-8859; Email: jinzhang@pku.edu.cn

Authors

Liu Qian – Center for Nanochemistry, Beijing Science and Engineering Center for Nanocarbons, Beijing National Laboratory for Molecular Sciences, College of Chemistry and Molecular Engineering, Peking University, Beijing 100871, People's Republic of China

Ying Xie – Center for Nanochemistry, Beijing Science and Engineering Center for Nanocarbons, Beijing National Laboratory for Molecular Sciences, College of Chemistry and Molecular Engineering, Peking University, Beijing 100871, People's Republic of China

Mingzhi Zou – Center for Nanochemistry, Beijing Science and Engineering Center for Nanocarbons, Beijing National Laboratory for Molecular Sciences, College of Chemistry and Molecular Engineering, Peking University, Beijing 100871, People's Republic of China

Complete contact information is available at:

<https://pubs.acs.org/10.1021/jacs.1c08554>

Author Contributions

The manuscript was written through contributions of all authors.

Notes

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