

## A review of the large-scale production of carbon nanotubes: The practice of nanoscale process engineering

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Carbon nanotubes (CNTs) are nanomaterials that have attracted great research interest because of their unique properties and promising applications. The controllable synthesis of CNTs is a precondition for their broad application. In this review, we consider nanoscale process engineering and assess recent progress in the mass production of ultra-long, inexpensive CNTs with good alignment as well as tunability in wall number and diameter for fundamental and engineering science applications across multiple scales. Cutting-edge nanoscale process engineering research in the areas of physics, chemistry, materials, engineering, ecology, and social science will allow us to obtain high added value and multi-functional advanced CNTs. The synthesis of CNTs with controllable chirality, good-alignment, and predetermined sizes and lengths still presents great challenges. Through multidisciplinary scientific research, advanced CNT-based materials will promote the development of a sustainable society.

**carbon nanotube, process engineering, mass production, synthesis, chemical vapor deposition, nanotechnology**

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Iijima's 1991 paper brought carbon nanotubes (CNTs) to the forefront of the scientific community [1]. CNTs can be multi-walled with a central tubule of nanometer diameter surrounded by graphitic layers separated by  $\sim 0.34$  nm. By contrast, single-walled CNTs (SWCNTs) are a cylindrical tube formed by wrapping a single-layer graphene sheet. According to the wall number of a CNT, it can be classified as a SWCNT, double-walled CNT (DWCNT), triple-walled CNT (TWCNT), or multi-walled CNT (MWCNT). Because of the covalent  $sp^2$  bonds between individual carbon atoms, a nanotube can have a Young's modulus between 1.2–5.5 TPa, a tensile strength about a hundred times greater than that of steel, and can tolerate large strains before mechanical failure. A CNT can be a metal, semiconductor, or small-gap semiconductor. The state of the CNT depends heavily on the (m, n) indices, and, therefore, on the diameter and chirality. CNTs also possess tunable surfaces characteristics,

well-defined hollow interiors, and high biocompatibility with living systems. Based on these properties, many potential applications, including both large-volume applications (such as conductive, electromagnetic, microwave absorbing, high-strength composites; super capacitor, or battery electrodes; catalyst and catalyst support; field emission displays; and transparent conducting films) and limited-volume applications (such as scanning probe tips; drug delivery systems; electronic devices; sensors; and actuators) have been proposed [2].

Controllable mass production of CNTs with the desired structures and properties is essential for future applications. In the past 20 years, arc discharge, laser ablation, and chemical vapor deposition (CVD) methods have been developed to produce CNTs in sizeable quantities. In CVD methods, the catalytic decomposition of a carbon feedstock into carbon and hydrogen atoms is initiated on an active catalyst surface, where the tubular CNTs grown. CVD growth can be achieved under mild conditions (such as normal pressure

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and low growth temperature) at a low cost. The wall number, diameter, length, and alignment of these CNTs can be well mediated. Thus, the CVD method is the most promising method for the mass production of CNTs.

Revolutionary discoveries in nanoscale science and technology over the past few decades have advanced nanoscale process engineering (NPE), which mainly deals with the transformation of materials and energy into nanostructured materials and nanodevices [3]. NPE synergizes the multiple disciplines of physics, chemistry, materials science and technology, biotechnology, and information technology. The CNT production process many different time and length scales [4]. On the microscopic scale, the carbon atoms from the decomposition of the carbon source self-assemble into a one-dimensional (1D) tubular CNT structure with the help of a catalyst. On the macroscopic scale, the continuous mass production of tons of chemical product occurs. This process is similar to other modern processing industry techniques, including processing for high-purity MWCNTs and SWCNTs, and the processes of heat and mass transfer. Great efforts have been devoted to CVD growth to obtain CNTs with desired structures. For large-volume applications, inexpensive CNTs with good dispersion are prioritized; however, for small-volume applications, CNTs with controllable lengths, orientation, and chirality are required.

Many other articles have reviewed the growth of single- and multi-walled carbon nanotubes, using either arc discharge, laser ablation, or catalytic CVD techniques [4–16].

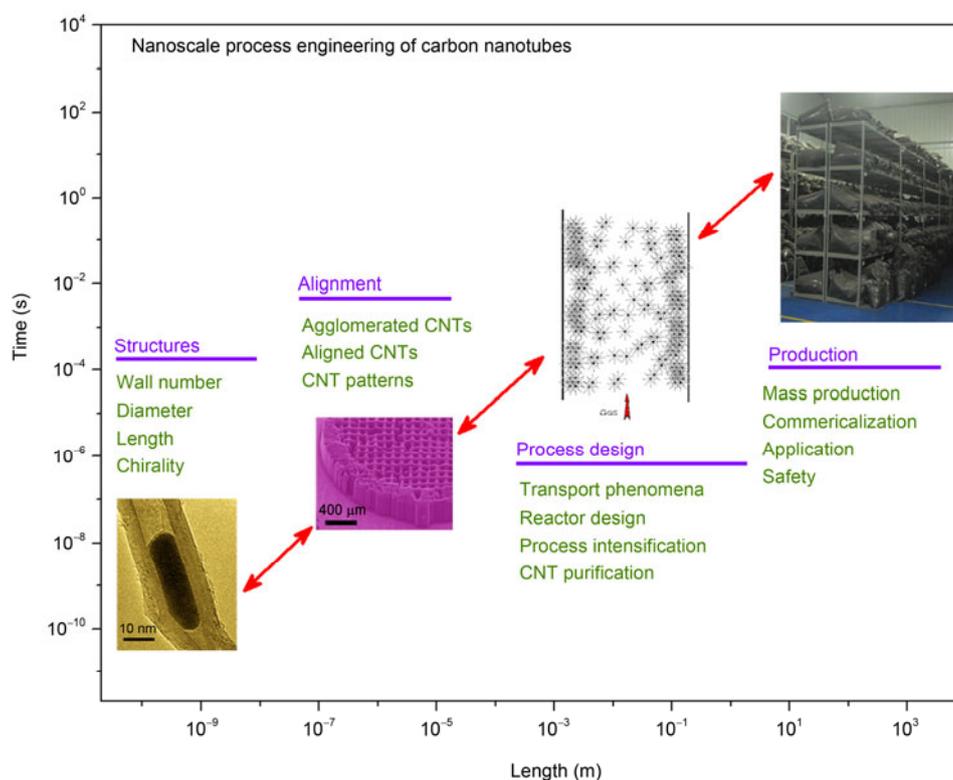
Herein, our objective is to shed light on the recent progress of controllable growth of CNTs. The recent progress on the synthesis of wall number and diameter tunable CNTs, super-long CNTs, aligned CNTs, high-purity CNTs, and cheap, mass-produced CNTs based on our recent research are reviewed, which presents a typical pioneer model to for the practice of NPE.

## 1 Wall number and diameter-mediated CNT production

The mechanical, electronic, and optical properties of CNTs depend on the wall number and diameter of a CNT. Simple and scalable methods for wall number and diameter-mediated CNT production are critical. Controlling the state of catalyst particles and tuning the atmosphere in the growth of CNTs are two typical strategies.

The size of the catalyst particles has an important role in the diameter of the CNTs. Usually, small catalyst particles (0.5–5 nm) are efficient for SWCNT growth, and large catalyst particles (8–100 nm) often produce MWCNTs. However, metal nanoparticles are prone to aggregate into larger ones at high temperatures during the growth of CNTs. Great effort has been devoted to both controlling the initial sizes of catalysts and preventing the particles from sintering.

Taking powder catalysts as an example, it has been demonstrated that the loading amount of the active phase



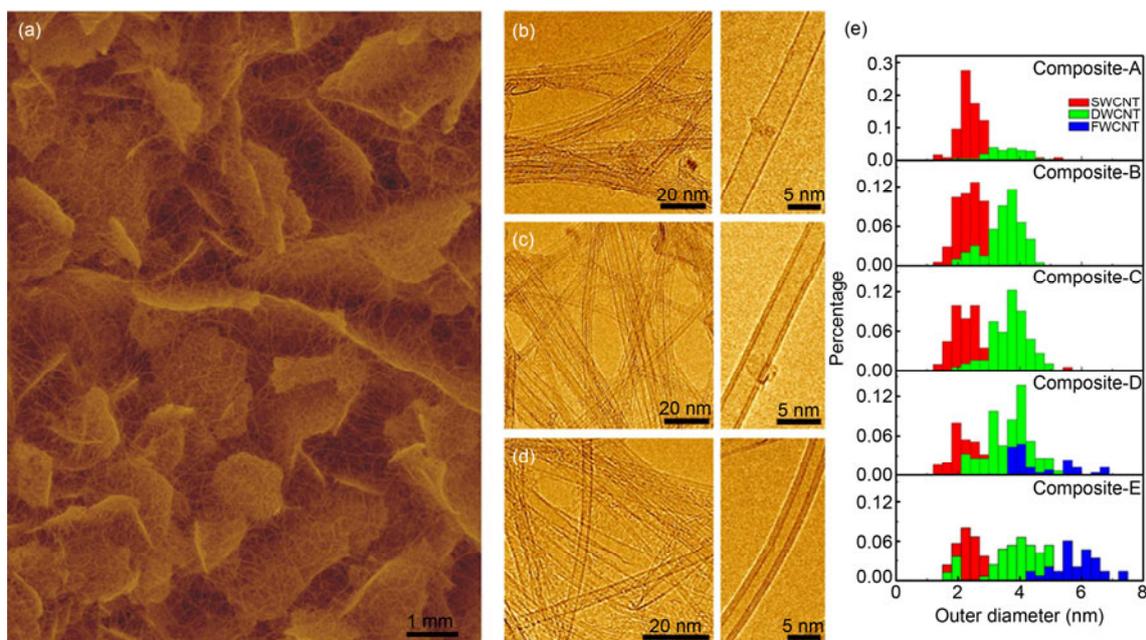
**Figure 1** Multi-scale illustration of the nanoscale process engineering of CNTs.

(consequently, the different catalyst particle sizes) significantly influences the wall number and diameter of CNT products. When the loading amount of Fe on MgO was increased, discrete catalytic nanoparticles with diameters in the range of 1–2 and 3–5 nm supported the growth of SWCNTs with a diameter distribution of 0.9–2.7 and 1.5–6.0 nm, respectively. The further increases in Fe content caused the formation of SWCNTs with larger diameters, as well as DWCNTs. Therefore, it is important to tune the size of metal particles by selecting proper catalyst precursors or carefully controlling the catalyst formation process. In the thermal CVD for the synthesis of aligned CNTs, precisely controlled catalyst layers with different thicknesses are commonly employed to modulate the size of catalyst particles and the structure of CNTs. Generally, CNTs synthesized on thinner catalyst films have smaller wall numbers. Deposition through nano-templates can also control the catalyst sizes, leading to the preferential growth of wall-number-selected CNTs.

To reduce the influence of catalyst sintering, a physical barrier, such as a surfactant, ligand, dendrimer, an open shell of support, or a meso/nanoporous oxide shell, have been introduced to separate metal nanoparticles from each other to prevent them from sintering. Several chemical approaches which depend on the formation of alloy hybrid materials or substrate effects have been explored as ways to stabilize the metal nanoparticles. Recently, we demonstrated that high-density metal nanoparticles (ranging from  $10^{14}$  to  $10^{15} \text{ m}^{-2}$ ) can be formed on flakes of layered double hydroxides (LDHs), a class of synthetic two-dimensional (2D) nanostructured anionic clays consisting of positively charged

layers with charge-balancing anions in between them. Thus, the metal cations are dispersed at an atomic level in synthesized LDHs, which can be reduced into well-dispersed and thermal-stable nanoparticles because of the strong interaction between the metal nanoparticles and the calcined LDH flakes. Because of the suppression in catalyst sintering, high-yield SWCNTs can be selectively synthesized on the Fe/Mg/Al (0.05:1.85:1) LDHs, and the tube distribution can be easily mediated by changing the Fe amount (Figure 2) [17]. With increasing iron content in LDH catalysts, more DWCNTs and MWCNTs are found. It can be directly observed that particles smaller than 3 nm are more inclined to form SWCNTs, and that DWCNTs usually grow from particles with a size of 3–5 nm. By contrast, for particles larger than 5 nm, the formation of MWCNTs encapsulated particles and led to catalyst deactivation (Figure 2(b)–(e)). We also explored the idea of chemically-mediated-precursor-induced formation of metal nanoparticles with higher density (ranging from  $10^{15}$  to  $10^{16} \text{ m}^{-2}$ ) on  $\text{MoO}_4^{2-}$  intercalated LDH flakes [18,19]. The metal nanoparticles were much smaller and had better thermal stability because of the pinning effect, and the Mo-containing LDH flakes were demonstrated to be excellent catalyst precursors for the production of aligned SWCNTs.

On the other aspect, a carefully controlled synthesis process can also tune the fine structure of CNTs. High temperature provides the energy for the reorganization of carbon atoms into CNTs, and is one of the main parameters in CNT synthesis. Controlling the temperature in the annealing process of the catalyst precursor benefits the formation of the desired catalyst particles for CNTs. Lowering the growth

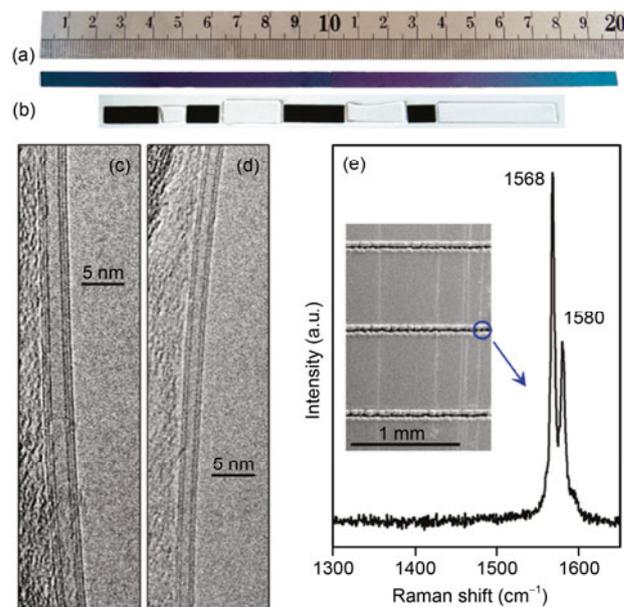


**Figure 2** (a) SEM image of CNTs synthesized on LDH flakes; (b)–(d) TEM and high-resolution TEM images of CNTs grown on LDH flakes with different Fe loading amounts; (e) CNT diameter and wall number on CNTs grown on LDHs (Fe loading amount: A, 1.7%; B, 3.8%; C, 6.2%; D, 10.8%; E, 24.1%) [17].

temperature has also been found to be an efficient way to reduce CNT diameter in the floating catalyst process [20]. Another important method by which to tune the CNT structure is adding elements that can affect the growth process. Using liquefied petroleum gas (LPG, containing some sulfur) as a carbon source can reduce the diameter of CNTs, most likely because of the partial poisoning of the catalyst particles [21]. Thiophene has also been widely used as an additive to promote the formation of SWCNTs in the floating catalyst process for similar reasons [22]. The introduction of weak oxidants (such as H<sub>2</sub>O and CO<sub>2</sub>) during the growth process is an efficient method to mediate the CNT structure. For instance, when CO<sub>2</sub> was introduced, SWCNTs or DWCNTs were selectively synthesized [23]. When the CO<sub>2</sub> concentration increased from 0 to 36.8%, the carbonaceous impurities were eliminated and the ratio of SWCNTs increased from 20% to 70%. It is possible that the effects of CO<sub>2</sub> in the process generate H<sub>2</sub>O and also serve as a weak oxidant that modulates the structure of CNTs.

## 2 Super-long CNT production

Super-long CNTs are fascinating materials because they allow for the easy fabrication of multiple field-effect transistor devices using an individual tube and offer the potential for direct dry-spinning into a super-high-strength fiber. To date, the only effective method to prepare these materials is with gas-flow-directed CVD on a Si substrate [9,10,24,25]. In this method, catalyst particles catalyze the decomposition of the carbon source to provide carbon atoms and the gas flow provides the buoyancy for suspended tubes to grow without resistance. This method shows a growth rate of 10–24 μm/s (with a reported rate as high as 50 μm/s). Water served to maintain the catalyst activity during the CVD growth of super-long CNTs. With optimized water feed concentration, the average growth rate of 10–20 cm CNTs increased from 20 to 80–90 μm/s. A 20-cm long substrate was obtained by connecting two 10-cm long Si substrates (each with a 700-nm thick SiO<sub>2</sub> layer on the surface, Figure 3(a)) or by connecting many short Si and SiO<sub>2</sub> substrates in sequence (Figure 3(b)) [26]. A high-resolution TEM revealed that the CNTs longer than 3 cm were mostly TWCNTs (with diameters of about 3 to 5 nm and selectivity >90%) in the reaction in which water was not added, and were a mixture of 49% DWCNTs (outer diameter of 1.7–2.0 nm), 40% TWCNTs (outer diameter of 2.5–3.3 nm), and 11% SWCNTs (outer diameter of 2.4–3.3 nm) when water was introduced (Figure 3(c) and (d)). The Raman spectra of a DWCNT growing across the trenches (cut by a laser on the Si substrate and about 100 μm wide, as shown in the inner picture in Figure 3(e)) showed no D band but did show a very strong G band. The G band included a strong peak at 1568 cm<sup>-1</sup> and a relatively weak peak at 1580 cm<sup>-1</sup>. This result indicates that the long tubes with nearly



**Figure 3** (Color online) (a) Optical microscope image of the 20-cm long Si substrate and (b) the substrate connected through Si (dark) and SiO<sub>2</sub> (white) substrates for 20-cm long CNT growth. (c), (d) TEM images of DWCNT and TWCNT. (e) Raman spectra of DWCNT [26].

perfect structure would be ideal for the fabrication of quantum conductive devices. The water removed the amorphous carbon that covered the active metal catalyst in the CVD method. The replacement of part of the Si substrate with SiO<sub>2</sub> substrates resolves the length limitation set by the substrates and also decreases the manufacturing cost of centimeter-scale DWCNTs and TWCNTs.

## 3 Aligned CNT production

Aligned CNTs possess many attractive properties, such as identical tube length, uniform orientation, and ultra-high purity. Compared with agglomerated S/D/MWCNTs, it is much easier to fabricate different CNT macroscopic structures (such as fluffy CNTs [27], CNT fibers [28–30], and CNT films [30]) with aligned CNTs. They also exhibit excellent performance when used as filler for polymer reinforcement to improve the electronic, mechanical, and thermal properties of polymers [31]. Apparently, exploration of the growth mechanism and the realization of production for aligned CNTs have stimulated their applications and commercial uses.

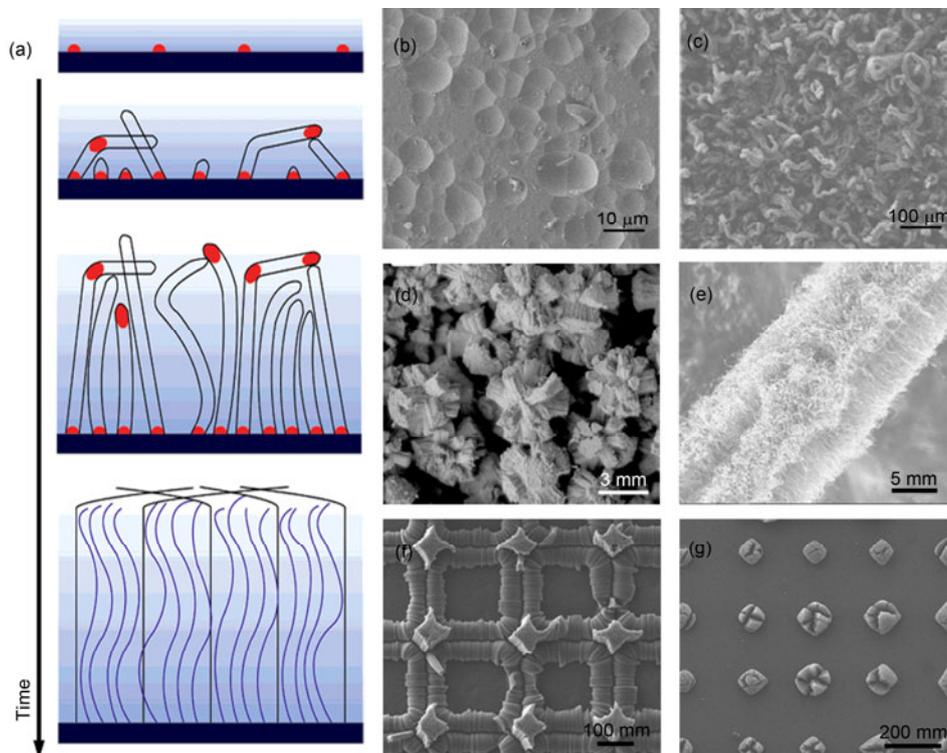
In the last few decades, most research on the growth mechanism has focused on the atomic-scale growth of an individual CNT, such as the vapor-liquid-solid model. However, this research has not sufficiently explained the interaction between CNTs and the mechanism of the formation of different CNT agglomerates. In fact, the formation of the initial woven structure of CNTs has a critical role in the formation of aligned CNTs [32]. During the

initial CVD growth, CNTs grew in random directions and interlaced with each other to form a uniform woven structure, in which neighboring CNTs were entangled because of van der Waals forces. Subsequently, CNTs were not able to grow freely on the surface because of horizontal and vertical space resistance. They had to grow under the woven structure. Stress was thus created by the interactions between upward-growing curved CNTs that formed later in the process and straight CNTs that formed earlier in the process, and which composed the woven structure (Figure 4(a)). The stresses kept the straight and curved CNTs growing at the same macroscopic rate. The Onsager virial theory predicts a phase transition from a random distribution to an ordered distribution above a transition value of  $D/(vL)$ , where  $D$  and  $L$  are the diameter and length of the rod-like particles, respectively, and  $v$  is the volume concentration [37]. When  $D/(vL)$  is less than 3.3, the distribution is more stable. The opposite occurs when  $D/(vL)$  is greater than 4.8. When the CNTs are short, the distribution is more stable. However, for the array, a transition from random to order alignment occurred; thus, the self-organization of the aligned CNTs exhibited a long length and a high yield. In practice, using different substrate morphologies can give rise to different types of CNT structures because of the initial formation of the top woven structures [33]. For flat substrates, uniform and flat CNT woven structures form over a large area during the growth of CNTs and give rise to

large-area, aligned CNTs with the same height. For rough substrates with a 10- $\mu\text{m}$  round concave structure, the woven structures formed separately in different concave structures and did not interact with each other. Therefore, single CNT ropes (small bundles of aligned CNTs) grew from the concave structures (Figure 4(b) and (c)). By contrast, ultra-rough substrates with nanoscale gaps (around 100 nm) failed to support the formation of a CNT woven structure on the top, and highly entangled CNTs agglomerates were formed.

The formation of aligned CNTs requires a relatively small area as a substrate to support their growth. By selecting different substrates and carefully modulating the interactions among CNTs, various 3D nanoarchitectures can be fabricated, which demonstrate fascinating performance and have advanced applications. Radial aligned CNT/spheres (Figure 4(d)) [34,38,39] and CNT/fiber hybrids (Figure 4(e)) [40,41] can be easily fabricated when spheres or fibers are employed as substrates. Levorotatory or dextrorotatory CNT arrays were twisted *in situ* into a double-helix can on a single calcined-layered double-hydroxide flake [42].

Different aligned CNT patterns, which show superior performance as field emission devices [43] and superhydrophobic materials [36], were constructed. One conventional method for patterning aligned CNTs is the pre-patterning of catalyst layers. Typical methods include lithography followed by e-beam evaporation of catalysts [44], selective wettability control followed by dip coating [45], and the soft contact



**Figure 4** (a) Schematic diagram of the synchronous growth of aligned CNTs [32]; (b) SEM image of a rough substrate with 10- $\mu\text{m}$  concave structures; and (c) CNT ropes synthesized on this substrate [33]; aligned CNTs grown on (d) ceramic spheres [34] and (e) fibers [35]; (f)–(g) CNT architectures grown using a pattern transfer method [36].

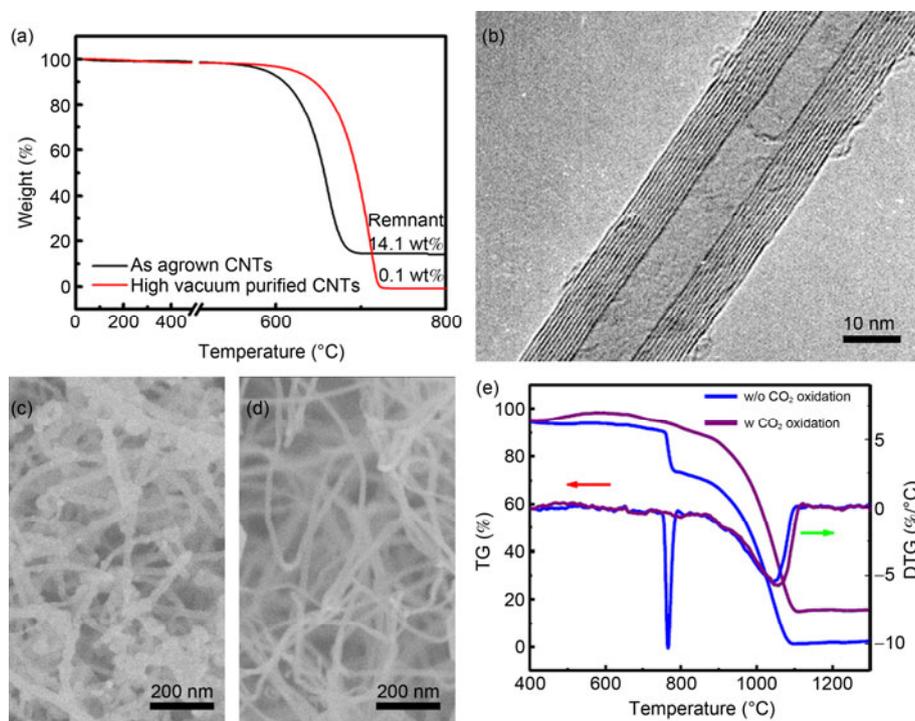
imprint method [46]. Such methods can be used to carefully control the structure at the micron scale. By selectively sacrificing the catalysts in the area without barrier layers, the patterning of aligned CNTs was realized [47]. As an alternative route, the patterning of aligned CNTs can also be realized during the growth process instead of through the pre-patterning of catalysts or substrates. 3D CNT architectures with patterned morphologies have been fabricated by a thermal pattern transfer method [42], in which the components of the masks served as promoters/inhibitors to increase/decrease the catalyst activity for the self-organization of CNTs into a family of patterned nanoarchitectures (Figure 4(f) and (g)). These methods provide a convenient way to make simple aligned CNT architectures.

#### 4 High-purity CNT production

CNTs prepared through a powder catalyst using the above methods inevitably contain metal catalyst nanoparticles, catalyst support materials, and carbonaceous impurities, which are obstacles to many applications. Metal impurities are usually residues from the transition metal catalysts. Some of the metal particles are encapsulated by carbon layers, making them impervious to acid treatment. Another problem that must be overcome is that carbonaceous and metal impurities have very wide particle size distributions and different numbers of defects and curvatures depending on

synthesis conditions. An overview of the purification of CNTs [48], including chemical oxidation, physical separation, and combinations of chemical and physical techniques, was described recently. We found that physical separation methods, such as ball milling, shearing, and ultrasonic treatment, can only break up large CNT agglomerates into small agglomerates [49]. A combined method was developed for the effective purification of agglomerated CNTs. A chemical treatment with a 3:1 concentrated  $\text{H}_2\text{SO}_4/\text{HNO}_3$  mixture gives a good dispersion by effectively severing the entangled CNTs. After annealing at  $2000^\circ\text{C}$  and  $0.01\text{--}0.1$  Pa for 5 h, CNTs of 99.9% purity with good graphene layers were obtained (Figure 5(a) and (b)) [50]. By contrast, raw DWCNTs produced with a fluidized bed reactor were purified efficiently by dissolving MgO and Si in HCl and HF solutions, sequentially. The residual Fe particles were removed through evaporation at  $1700^\circ\text{C}$ . At this temperature, DWCNTs with 99.25% carbon content and 85% yield can be achieved [51]. Recently, a self-sacrificed powder catalyst was developed and the carbon yield reached  $179 \text{ g}_{\text{CNT}}/\text{g}_{\text{catalyst}}$  [52]. In other words, the impurity content was suppressed to 0.55 wt%. No further purification is needed for many applications.

The synthesis of aligned CNTs faces similar problems regarding product purity. The aligned CNTs will not blend with support substrates when they grow on a wafer, but the amorphous carbon and metallic catalysts still remain as impurities, especially during the floating catalyst process. During



**Figure 5** (a) TGA curve for the high-purity CNT product and high-vacuum purified CNTs, showing the remnant reduced from 14.1 wt% to 0.1 wt%; (b) a high resolution TEM image of the purified CNT; (c) and (d) show the SEM images of CNTs without and with weak oxidation purification; (e) the  $\text{CO}_2$  atmosphere TGA curves of a CNT sample with and without  $\text{CO}_2$  oxidation, showing the elimination of amorphous carbon through the oxidation process [53].

CVD growth, thermal pyrolysis occurs in the gas phase along with catalytic decomposition [54]. This undesirable side effect can cause the continuous accumulation of amorphous carbon on the top surface and side walls of the aligned CNTs. The use of some weak oxidants in the growth process is helpful in suppressing the formation of amorphous carbon. Water is a commonly used promoter, which can eliminate the amorphous carbon while accelerating the growth of aligned CNTs [55]. CO<sub>2</sub> also improves CNT purity during high temperature growth (Figure 5(c) and (d)) [23,53,56,57].

## 5 Low-cost and large-scale CNT production

The above sections discussing the modulation of CNT structures and sample purities are mainly realized within the laboratory, and cannot satisfy the demands of industrial applications. For widespread use of a material, large-scale production at a low cost is a basic requirement. Exploring ways to lower the cost of production and developing an engineering method to handle CNTs are prerequisites for commercial application of this technology.

In the traditional chemical industry process, 60%–90% of production costs come from raw materials. It is important to select proper feedstock (such as catalyst supports, catalysts, and carbon sources) for the efficient production of CNTs at a low cost. Investigations into new, cheaper feedstocks, and more efficient catalyst/support combinations suitable for the mass production of agglomerated and aligned CNTs with a high yield are needed. Growth of CNTs on natural materials has great potential to achieve environmentally benign, low-cost production [58]. Some minerals, such as garnet sand, wollastonites [41], montmorillonite, vermiculite [59], sepiolite [60], and biomass-based activated carbon, have been used as catalysts and/or catalyst supports for the synthesis of CNTs. Organic natural materials, such as coal, natural gas, liquefied petroleum gas [21], camphor, eucalyptus oil, turpentine oil, and city gas can serve as carbon sources for CNT synthesis [58]. Various process intensification technologies, such as advanced catalysis processing, multi-functional reactors (such as two-stage fluidized beds), coupled-process development, and one-step synthesis routes for direct CNT applications have been proposed to efficiently produce CNTs [16]. However, exploring effective cheap feedstock to obtain CNTs with high purity, ideal structures, and high yields is still a challenge.

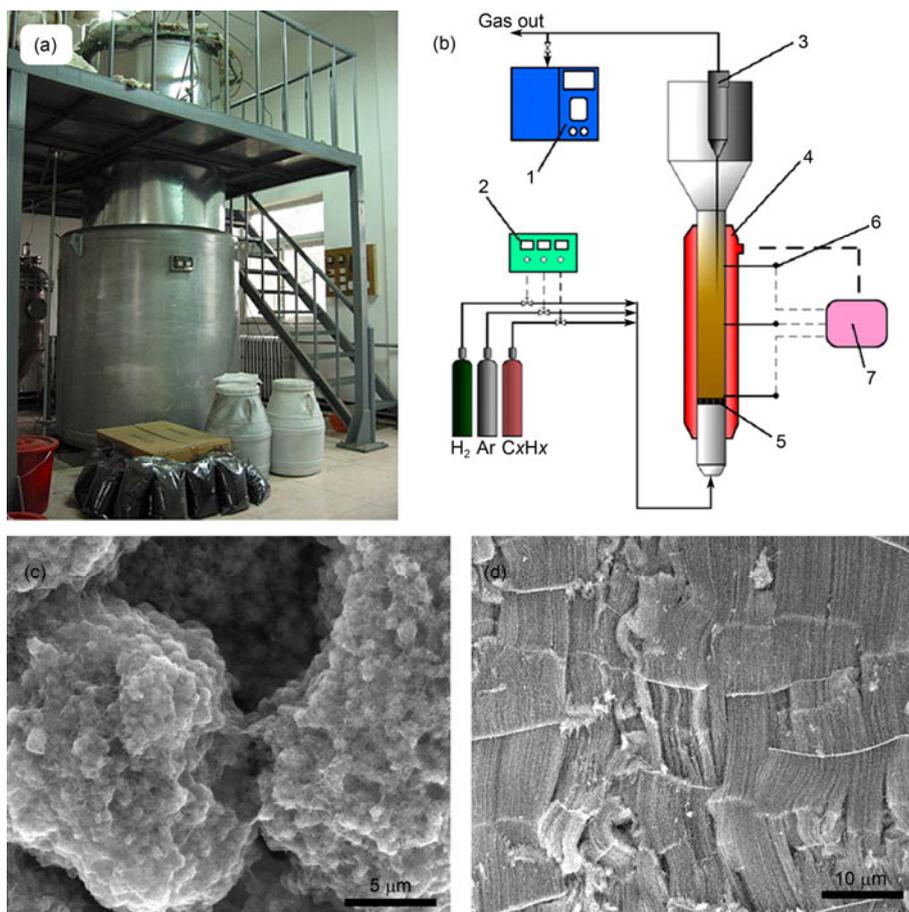
Mass production of CNTs has stimulated their applications and commercial uses. The most efficient way to mass produce CNTs is through fluidized bed catalytic CVD (Figure 6(a) and (b)) [4]. In a fluidized bed reactor, the solid particles (such as catalyst and its products) are transformed into a fluid-like state through suspension in a gas or liquid. The fluidized bed reactor has great advantages for CNT production, such as providing sufficient growth space,

excellent diffusion and heat transfer, ease in scaling up, and continuous operation. The CNTs grown on powder catalysts became entangled and formed agglomerates with a size of 10–200 μm, which are A particles, according to the Geldart particle classification. The powders in Group A exhibit dense phase expansion at a minimal fluidization velocity, and this state can be maintained over a large velocity range. Agglomerated MWCNTs (Figure 6(c)) [4,62], DWCNTs [63], and SWCNTs [64] have been efficiently synthesized in a fluidized bed reactor. A pilot mass-production project of agglomerated MWCNTs with high yields (15 kg/h) was realized in a fluidized bed reactor in 2002 [62]. For MWCNT production, the producers are mainly located in China, Japan, the United States, Germany, and Korea. Based on the fluidized bed technologies developed at Tsinghua University, the commercial production of MWCNTs had been achieved by Cnano Technology located at Beijing in China with a capacity of 560 tons per year as of the middle of 2009 [16]. With regard to S/DWCNT production, both the CVD and the arc-discharge methods are used by industrial producers.

Most aligned CNTs are synthesized on a flat surface. The surface area of the flat substrate is often limited and its mobility is poor. With flat silica as the substrate, the production of aligned CNTs is only about 1 g/h. If a substrate with a larger surface area is used, more aligned CNTs can be produced. Small spherical particles have large surface areas. These spheres also exhibit good flowability and can be transferred easily in the reactor. Radial growth on spheres is a good strategy for the mass production of aligned CNTs [35]. However, the spherical substrates used in our research had a size of about 0.6–1.2 mm, and are D particles, according to the Geldart particle classification. They can be fluidized in spouted beds with strong collisions, which may disturb the growth of the CNT array in a fluidized state. If the CNTs can be grown within a single particle, then the collisions among CNT arrays during growth can be avoided. Lamellar vermiculite catalyst particles with a size of 100–300 μm obtained using ion-exchange methods were used as a catalyst for the intercalated growth of aligned CNTs [61,65]. Both the catalyst and the product have a size of 80–200 μm and can be easily fluidized. An output of 3.0 kg/h aligned CNTs was realized in a pilot study in a plant fluidized bed reactor (Figure 6(d)) [65], which produced a large amount of CNT arrays for applications in the future.

## 6 Conclusions and remarks

Widespread investigations of the properties of CNTs, an advanced manmade material, and their large-scale application will only be possible when good quantities of specific types of CNTs can be produced. Much progress has been made in the mass production of ultra-long inexpensive CNTs with tunable wall numbers and diameters as well as



**Figure 6** (a) Pilot fluidized bed reactor [4] and (b) schematic diagram of a fluidized bed reactor [61]: 1, gas chromatogram; 2, mass flow controller; 3, cyclone; 4, reactor; 5, gas distributor; 6, thermocouple; 7, heat controller; SEM image of the mass-produced (c) agglomerated CNTs [4]; and (d) aligned CNTs [59].

good alignment for fundamental and engineering science applications across multiple time and length scales. This paper discussed the use of NPE for advanced nanostructured material production. Cutting-edge research in the areas of physics, chemistry, materials, engineering, ecology, and social science provides a great impetus for engineering practices to achieve high added-value and multi-functional advanced materials (such as CNTs).

However, compared with traditional bulk chemicals, the controllable synthesis and large-scale applications (on the order of millions of tons) of CNTs is still far from being achieved because of the difficulty of both synthesis and the subsequent product treatments. The synthesis of CNTs with controllable chirality, super-alignment, and predetermined size and length still presents great challenges. Furthermore, the strong coupling among CNT structures, production processes, properties, post-treatments, and applications still present difficulties. Environment, health, safety, and ecological considerations for CNTs also must be considered to ensure the development of intrinsically safe, high efficiency, and economical processes. Developing a means to produce these products through an integrated NPE method capable

of producing CNTs with all the desired characteristics is necessary.

With the great effort of the science and engineering community since Iijima's landmark 1991 paper, agglomerated S/MWCNTs and aligned MWCNTs have already been mass produced in orders of several to several tens of kilograms per hour; some have been used in Li ion batteries and advanced nanocomposites. The CNT is an extraordinary structure that demonstrates the power of nanotechnology. The mass production and applications of CNTs demonstrate the power of NPE. With multidisciplinary scientific research, NPE will provide advanced CNT-based materials that will help us to develop a sustainable society.

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