

## Review Article

# Synthesis, Properties, and Applications of Low-Dimensional Carbon-Related Nanomaterials

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In recent years, many theoretical and experimental studies have been carried out to develop one of the most interesting aspects of the science and nanotechnology which is called carbon-related nanomaterials. The goal of this paper is to provide a review of some of the most exciting and important developments in the synthesis, properties, and applications of low-dimensional carbon nanomaterials. Carbon nanomaterials are formed in various structural features using several different processing methods. The synthesis techniques used to produce specific kinds of low-dimensional carbon nanomaterials such as zero-dimensional carbon nanomaterials (including fullerene, carbon-encapsulated metal nanoparticles, nanodiamond, and onion-like carbons), one-dimensional carbon nanomaterials (including carbon nanofibers and carbon nanotubes), and two-dimensional carbon nanomaterials (including graphene and carbon nanowalls) are discussed in this paper. Subsequently, the paper deals with an overview of the properties of the mainly important products as well as some important applications and the future outlooks of these advanced nanomaterials.

## 1. Introduction

Carbon (from the Latin language, Carbo means coal) as a non-metal element can be found in every living organism; therefore, it can be bravely confirmed that the basis of one aspect of life is carbon. Carbon, as the original component presented in millions of various compounds, is capable to be very hard such as diamond or very soft as graphite. The use of this unique chemical element is almost unlimited from the hardest diamond for drilling to the softest form, graphite, for using as a lubricant in skin health and beauty. This nontoxic element is used as a filter to reduce other toxins. It has the highest melting point among all the known elements and occurs free in nature. On the other hand, carbon nanomaterials are known as the chemical fruits of the mother tree of carbon and most importantly carbon 60 molecules. Since a series of exciting carbon nanomaterials are arising, they have attracted tremendous attention and have been intensively studied on numerous carbon-based nanomaterials because of their unique structure, electronic, mechanical,

optical, and chemical characteristics. Low-dimensional carbon nanomaterials can be divided into categories of different dimensionality ranging from zero-dimensional (0-D) to one-dimensional (1-D) and two-dimensional (2-D) depending on their nanoscale range (<100 nm) in different spatial directions. The representatives in family of low-dimensional carbon nanomaterials focus on fullerene, onion-like carbon, carbon-encapsulated metal nanoparticles, nanodiamond (0-D), carbon nanofibers, carbon nanotubes (1-D), graphene, and carbon nanowall (2-D).

As a brief history, it is worth to remember that in 1952 Radushkevich and Lukyanovich introduced the hollow graphitic carbon fibers with 50 nm in diameter in the Soviet Journal of Physical Chemistry mentioned in [1]. In 1960, Bollmann and Spreadborough [2] showed the structure of multiwall carbon nanotubes (MWNTs) using an electron microscope. They investigated the friction properties of carbon due to rolling sheets of graphene in "Nature" [2]. In 1976, Oberlin et al. [3] showed the CVD growth of nanometer-scale carbon fibers. In 1985, the revolutionary

discovery in this area occurred, which was the discovery of fullerenes by Kroto et al. [4]. The discovery of fullerene seems to be very important because it might be the first new allotrope of carbon to be discovered in the 20th century [5]. The nickname for fullerene comes from Richard Buckminster “Bucky” Fuller (July 12, 1895–July 1, 1983), an American architect, author, designer, inventor, and futurist who used geodesic spheres structures in his work. Other important well-known carbon nanomaterials are carbon nanotubes (CNTs) which will be introduced with the full features and aspects in this paper. As a pictorial introduction, we have summarized the main structures of various low-dimensional carbon nanomaterials in Figure 1 [6–11].

In the past decade, by using nanotechnology and carbon-based nanomaterials, the world might be able to see an industrial revolution surpassing any one before. This new technology could end the world’s hunger, make affordable goods, have massive implications for medical breakthroughs, and unfortunately also be used in military applications. In this paper, we will discuss the synthesis, properties, and applications of these characteristic examples as well as the latest research results and developments, that hopefully more researchers can address the area and look forward to more research results.

## 2. Carbon Nanomaterials Synthesis

In this paper, our purpose is to summarize the well-known methods and to provide recent progress synthesizes of the entire nanostructure class of carbon nanomaterials. Where appropriate, we also provide a short historical background and the structure description of carbon nanomaterials.

### 2.1. Zero-Dimensional Carbon Nanomaterials (0-DCNs)

**2.1.1. Fullerene.** Fullerenes are spherical, caged molecules with carbon atoms located at the corner of a polyhedral structure consisting of pentagons and hexagons. A spherical fullerene looks like a soccer ball and is often called “buckyball.” Fullerenes were named after Richard Buckminster Fuller, an architect known for the design of geodesic domes which resemble spherical fullerenes in appearance. In fact, fullerenes were discovered as an unexpected surprise during laser spectroscopy experiments in 1985, by researchers at Rice University. As mentioned in the Nobel Prize records at [12], the 1996 Nobel Prize in chemistry was awarded jointly to Robert F. Curl, Jr., Richard E. Smalley, and Sir Harold W. Kroto “for their discovery of fullerenes” [12]. The first method of production of fullerenes shown in Figure 2 by Kroto et al. in 1985 [4] used laser vaporization of carbon in an inert atmosphere in which microscopic amounts of fullerenes were produced. However, for the first time in 1990, the physicists Krätschmer et al. [13] produced isolable quantities of C60 by using an arc to vaporize graphite. The way was thus open for studying an entirely new branch of chemistry. Alekseyev and Dyuzhev [14] systemically discussed fullerene formation in an arc discharge and all the aspects of the problem from arc

discharge calculations to the immediate fullerene molecule assembly.

Fullerene chemistry has become a very hot research field in the last two decades. A variety of fullerene derivatives with unique properties were produced, and several techniques for producing them in greater volumes have been suggested. In 1991 and 1992, Howard et al. [15, 16] observed fullerenes C60 and C70 from benzene/oxygen flames and developed a method of synthesis of fullerenes in combustion. On the other hand, Xie et al. [17] synthesized fullerenes C60 and C70 via microwave plasma from chloroform at low-pressure argon atmosphere. The microwave plasma synthesis from chloroform opened a new way to large quantity and low-cost production of fullerenes, various perchlorinated intermediates of fullerenes, and the perchlorinated carbon clusters. Taylor et al. [18] introduced a synthesis of C60 and C70 by pyrolysis of naphthalene at 1,000°C. The advantages involve a continuous process that does not require rod replacement, and closed fullerene cages can be prepared from well-defined aromatic fragments. Koshio et al. [19] also used a method of fullerene by pyrolysis ragged single-wall carbon nanotubes treated by ultrasonication with an organic solvent followed by heating in oxygen gas. As a new event in 2009, Chen and Lou [20] reported that C60 can also be synthesized from the reduction CO<sub>2</sub> via metallic lithium or MgCO<sub>3</sub> at 700°C, ca. 100 MPa. Although the yield via the above method was low, this method could resolve the biggest contamination problem in the previous synthesis methods and could provide some new insights for the formation of C60.

Koprinarov et al. [21] reported fullerene structures (FSs), and FSs with incorporated iron atoms were obtained via DC arc discharge between carbon electrodes in Ar and ferrocene gas mixture ambient. The method supplied the electrode deposit growth with carbon from the ferrocene, which made fullerene creation easier and increased the product quantity. Richter et al. [22] made a detailed research on the process of fullerene formation in acetylene/oxygen/argon flat flames with adding chlorine and burning at low pressure. Recently, many chemical syntheses of fullerenes also have been reported in [23].

**2.1.2. Carbon-Encapsulated Metal Nanoparticles (CEMNPs).** Carbon-encapsulated metal (magnetic) nanoparticles (CEMNPs) represent a new class of Zero-dimensional carbon-metal composite nanomaterials. It is the shape of core-shell structure on the nanoscale. The polyhedral metallic core is entirely encapsulated by the multilayer-graphitized carbon shell. So, the carbon layers isolate the particles magnetically from external environment and protect them against corrosion and magnetic coupling between individual particles.

Since the first report on LaC<sub>2</sub> encapsulated within nanoscale polyhedral carbon particles in a carbon arc synthesized by Ruoff et al. [24] and Tomita [25] in 1993, carbon-encapsulated metal nanoparticles have received considerable attention because of their novel structures and obvious technological promise. Several groups have succeeded in encapsulating various materials into a hollow graphitic

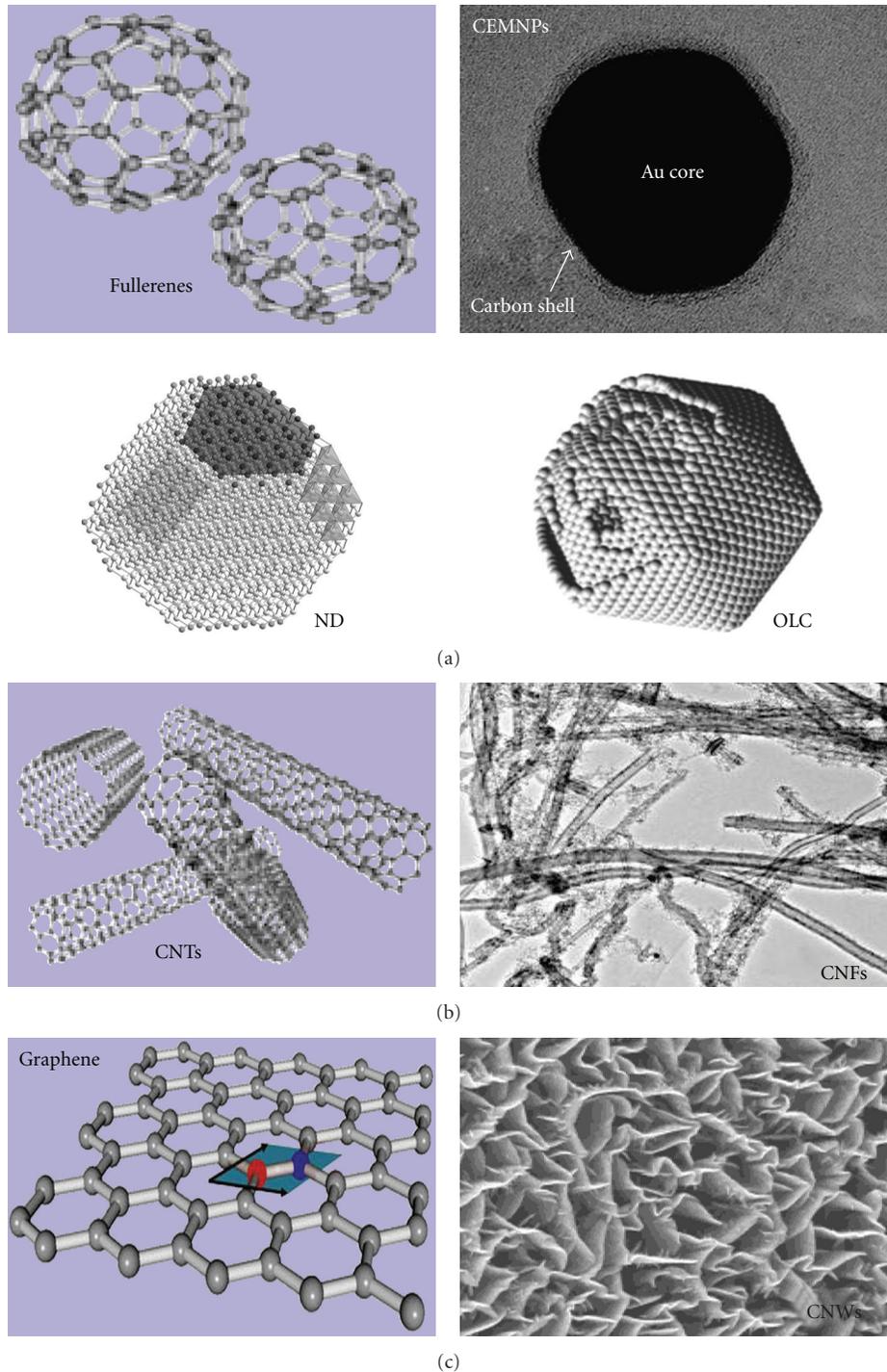


FIGURE 1: Crystal structures of the different low-dimensional nanocarbon. From left to right: (a) Fullerene, Carbon-encapsulated metal nanoparticles, Nanodiamond, and Onion-like carbon, (0-D); (b) carbon nanotube and carbon nanofibers, (1-D); (c) graphene and carbon nanowalls, (2-D) [6–11].

cage by arc discharge method. Saito [26] reported that 13 rare earth metals and iron-group metals were wrapped in graphitic carbon in 1995. Dravid et al. [27] solved the major problem about the production of large amounts of unwanted carbonaceous debris in the standard arc method with the synthesis of graphite-encapsulated nanocrystals.

They used tungsten arc technique which lowers the amount of carbonaceous debris produced by lowering the carbon content of the arc. Host et al. [28] also reported the structure and magnetic studies of carbon-coated nanocrystals of nickel and cobalt synthesized in a special low carbon to metal ratio arc chamber using tungsten arc techniques. Host et

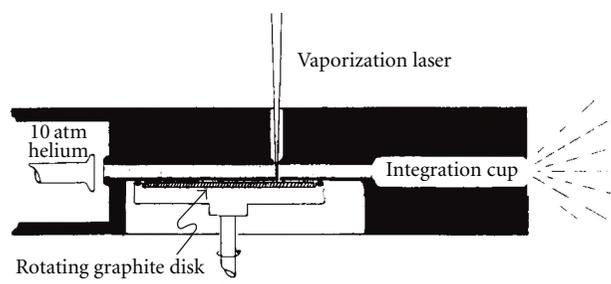


FIGURE 2: The schematic diagram of fullerene fabrication system using laser vaporization [4].

al. in 1998 [28] and Jiao et al. in 1996 and 1998 [29, 30] reported that a set of carbon encapsulated Ni, Co, Cu, and Ti particles were prepared by an arc discharge process modified in the geometry of the anode and the flow pattern of helium gas. However, the traditional method requires an expensive vacuum system to generate the arc plasma, and the carbon-encapsulated nanoparticles are only found in the soot deposited on the cathode. In 2004, Qiu and Tsang [31] reported the preparation of carbon-coated nickel metal nanoparticles using the arcing coal-based carbon rods submerged in deionized water. The coal-based carbon rods were prepared from a mixture of coal and nickel powders, and the arc discharge was carried out in water instead of in inert gases. The mentioned method resolved a variety of the previous problems.

In 1998, Harris and Tsang [32] described a new technique for carrying out the high-temperature heat treatments on microporous carbons used to encapsulate molybdenum, uranium, and cobalt. The method based on their researches solved the difficulties with the scaleup of low yield. In 2002, Flahaut et al. [33] reported cobalt nanoparticles encapsulated in graphitized carbon shells synthesized using catalytic chemical vapor deposition (CCVD) in high yield involving the reduction by an  $H_2/CH_4$  mixture of an  $Mg_{1-x}Co_xO$  solid solution prepared by the combustion method. In 2003, during the researches made by Wang et al. [34], carbon-coated cobalt nanocapsules were synthesized by the chemical vapor-condensation process with cobalt carbonyl ( $Co(CO)$ ) used as a precursor and carbon monoxide ( $CO$ ) as a carrier gas. Tsai et al. [35] reported that a method using microwave plasma enhanced chemical vapor deposition (MPECVD) system to synthesize pure carbon-encapsulated metal nanoparticles on silicon wafer without the existence of other carbon nanostructures.

Liu et al. demonstrated in their paper [36] that carbon-encapsulated cobalt nanoparticles were synthesized via the modified catalytic pyrolysis, the mechanical milling derived  $Co/NaCl$  as catalyst, and the productivity was almost 100%. This method is superior to early catalytic methods in high productivity and is fully separated from the supporting materials by simple washing process, especially the by-products carbon nanotubes (CNTs) [37].

In 2005, Lu et al. [38] reported the synthesis of carbon-encapsulated Fe nanoparticles via a picric acid-detonation-induced pyrolysis of ferrocene. Their technique was a

self-heating and extremely fast process. Wu et al. [39] reported a novel method for the synthesis of amorphous carbon-encapsulated  $Fe_7C_3$  nanocrystals via the explosion of a hybrid xerogel containing oxidized pitch and iron nitrate. Wang et al. [40] reported a continuous synthesis of high purity, high coercive force and good ferromagnetism CEMNPs based on the spray pyrolysis of a mixture of iron carbonyl and alcohol at  $500\text{--}900^\circ\text{C}$ . The technique based on spray pyrolysis offers a large-scale production, the simplicity of the apparatus, and good productivity. Song et al. [41] proposed that the similar carbon-encapsulated  $Fe_3C$  nanoparticles can be synthesized on a large scale via the cocarbonization of 1, 2, 4, 5-tetramethylbenzene and ferrocene under autogenous pressure. Huo et al. [42] synthesized Carbon-encapsulated iron nanoparticles with uniform diameters by cocarbonization of an aromatic heavy oil and ferrocene at  $480^\circ\text{C}$  under autogenous pressure. This preparation method of CEMNs is characterized by simplicity, low cost, controllability, and high yields. Bystrzejewski et al. [43] synthesized Fe,  $Fe_3C$ , and  $NdC_2$  carbon-encapsulated magnetic nanoparticles using thermal radio frequency (RF) plasma torch technique. In that paper, authors found the nonlinear transmission behavior in CEMNPs material with the smallest saturation.

In 2008, Park et al. [44] reported a method (pulsed laser irradiation synthesis, PLIS) to synthesize CEMNPs such as Fe-C, Ni-C, and Co-C by irradiating nanosecond laser pulses into a metallocene-xylene solution under room temperature and atmospheric pressure. There will be more CEMNP synthesis arising, because the PLIS method is simple and can be operated under ambient conditions. In 2006, Nishijo et al. [45] synthesized the carbon-encapsulated nanoparticles via thermal decomposition of metal acetylide at a low temperature. Using this method, nanoparticles of low-melting point Sn and metastable carbide phases of Pd, Ni, and Co were successfully encapsulated in amorphous carbon shells owing to the low synthesis temperature. Recently, Maya et al. [46] have produced and characterized encapsulated silver nanoparticles provided by high-current pulsed electric arcs system in an argon atmosphere. The main advantage of this method is that the metal nanoparticles are encapsulated in amorphous carbon from the beginning, and this layer acts as an efficient chemical barrier.

In summary, as briefed in Table 1, CEMNPs can be synthesized by a variety of techniques such as arc discharge method, tungsten arc techniques, high-temperature heat treatments, the mechanical milling, cocarbonization, pulsed laser irradiation, and high-current pulsed electric arcs system.

**2.1.3. Nanodiamond (ND).** As we all know, diamond is one of the carbon allotropes as graphite. Graphite is the most stable form of carbon at ambient pressure. Spherical and truncated octahedron diamond with predominant  $sp^3$ -bonded carbon is one of the hardest materials known to date and is often regarded as the king of all gemstone and top-drawer materials because of its excellent scientific qualities in hardness, chemical corrosiveness, thermal expansion and conductivity, electrical insulation, and biocompatibility. On

the other hand, nanodiamond (ND) is a cubic structural diamond. It possesses diamond structure and diamond properties. The average size is mere 5 nm in diameter. In the wide sense of the word, “nanodiamond” contains a variety of diamond-based materials at the nanoscale (the length scale of approximately 1–100 nm) including pure-phase diamond films, diamond particles, and their structural assemblies. Several synthesis methods have been developed to synthesize laboratory-produced nanodiamonds. There are two main methods for fabrication of nanodiamond: transformation of graphite under high temperature and high pressure and detonation of the carbon explosive materials.

In 1955, Bundy et al. [47] realized the 30-year dream of many scientists in which diamond can be transformed from graphite. Under the title “Man-made diamonds” in Nature [47], Bundy and his coworkers successfully reported the synthesis of diamond using a high-temperature and high-pressure process. However, the synthesis of diamond by the detonation of explosives with a negative oxygen balance in a steel container under vacuum condition was reported in the 1980s [48, 49]. There are also some related literatures in recent reports shown two mentioned methods [50–55]. Explosive detonation is still widely used; however, the process of the detonator explosion is extremely fast and very complex. Moreover, there are some disadvantages observed in detonation method. In fact, the fraction of surface to bulk atom and oxygen, hydrogen, and nitrogen content in the nanodiamond resulting from after-purification process are difficult to remove.

As an interesting matter, most previous researches on detonation synthesis have been done at military or commercial plants; thus several reports are available for the scientific community. Therefore, the best method is to develop new techniques to the synthesis of well-dispersed and pure nanodiamonds. Recently, more researches also about the aspects of low energy, low cost, easily controlled, few byproducts, controlled-sized, and large scale have been reported in related literatures, such as microwave plasma chemical vapor deposition [56–58], hot filament chemical vapor deposition [59], pulse laser ablation [60–62], electron irradiation [63], and high-energy X-ray diffraction [64].

**2.1.4. Onion-Like Carbons (OLCs).** Ugarte [65] in 1992 reported that carbon soot particles and tubular graphitic structures were radiated by intense electron-beam and reorganized into quasispherical particles. Subsequently, Harris and Tsang [66] in 1997 studied the structure of two typical nongraphitizing carbons by heat treatment. They observed the fullerene-like structure close to carbon nanoparticles. Then, a new model for nongraphitizing carbons was proposed which was different with the other representatives of the carbon family graphite, fullerenes, and nanotubes. The onion-like carbons (OLCs) have the three to eight closed graphitic shell structures with the hollow core. The outer diameters are in the range of 20–100 nm. The polyhedral nanoparticles exhibited a well aligned concentric and high degree of symmetry structure. Quasispherical shape, nanometer size, and surface specificity of OLCs have attracted enormous attention. Several routes were developed

from synthesis of carbon onions including arc discharge [67, 68], high-electron irradiation [69], chemical vapor deposition [70], radio frequency plasma [71] and high-dose carbon ion implantation into metals [72], and [73, 74] thermal annealing of diamond nanoparticles [75].

The current researches on OLCs are limited because of unmanageable reaction, many byproducts, complex equipments, and high cost. Extensive research has been devoted to search the optimal synthetic route. At present, the most OLCs were synthesized using vacuum annealing of nanodiamond particles at fixed temperatures [75]. For instance, in 2007, Bulusheva et al. [76] produced quasispherical and polyhedral OLCs using vacuum annealing of nanodiamond (ND) particles, and the first researched quantum chemistry characterization and the electronic structure of OLCs by X-ray absorption spectroscopy. Recently, in 2010, Bystrzejewski et al. [77] described a simple, facile, and low-cost process of the synthesis of OLCs by a catalyst-free ( $\text{NaN}_3\text{-C}_6\text{Cl}_6$  mixture) thermolysis route, and meanwhile, this method solved the problem of separation.

Zhao et al. [78] reported a large quantities synthesis of OLCs using carbonization of the solid-state catalyst of PF resin as the carbon resource and ferric nitrate as the precursor at 1000°C. Liu et al. [79] also synthesized OLCs using  $\text{Fe/Al}_2\text{O}_3$  as catalyst by chemical vapor deposition (CVD) at a relatively low temperature (400°C) and efficiently avoided the growth of CNTs. Du et al. [80] synthesized high-purity OLCs in high yields from coal by radio frequency plasma economically.

## 2.2. One-Dimensional Carbon Nanomaterials (1-DCNs)

**2.2.1. Carbon Nanofibers.** Carbon nanofibers (CNFs) are composed of stacked and curved graphene layers from a quasi-one-dimensional (1D) filament. CNFs have cylindrical or conical nanostructures. Their diameters vary from a few to hundred nanometers, while lengths range from less than a micrometer to millimeters. As shown in Figure 3, according to the angle between graphene layers and fiber axis, the morphological structure is often divided into plate CNFs, ribbon-like CNFs, herringbone CNFs [81].

CNFs known as filamentous carbon have been known for a long time [82]. However, the synthesis of filamentous carbons did not evoke great interest of scientists in those early years until the discovery of carbon nanotubes by Iijima in 1991 [83]. Generally, CNFs can be synthesized through the traditional vapor growth method [84–86], cocatalyst deoxidization process [84], catalytic combustion technique [85], plasma-enhanced chemical vapor deposition [87, 88], hot filament-assisted sputtering [89], ultrasonic spray pyrolysis [90], and ion beam irradiation [91].

The large-scale production of CNFs has usually been carried out by using PECVD in which CNFs are grown by catalytic decomposition of hydrocarbon under high temperature. Catalytic PECVD provides a means for the controlled synthesis of CNTs and CNFs and grows individual nanostructures with deterministic characteristics by changing the starting materials or plasma conditions during growth. This review will focus on the catalytic PECVD

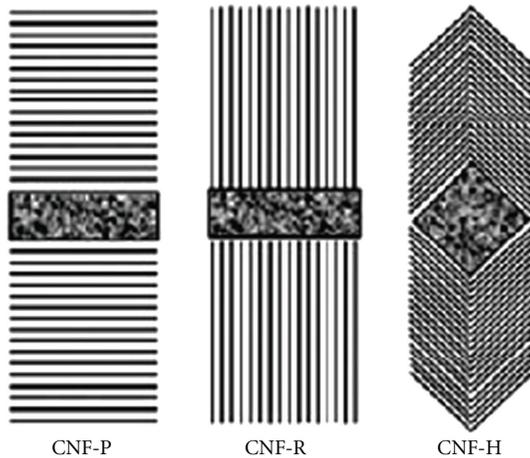


FIGURE 3: The schematic diagram of three different CNFs: platet CNFs and ribbon-like CNFs, herringbone CNFs [81].

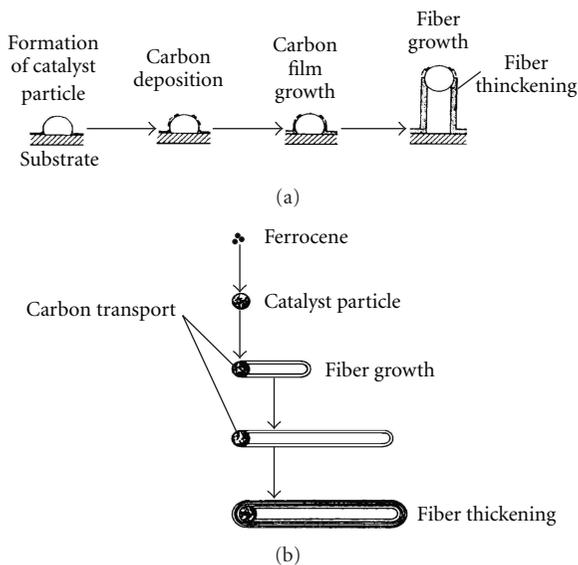


FIGURE 4: Two methods of synthesizing VACNFs: (a), “seeded catalyst on a flat plate” and (b), “floating catalyst” [92].

growth process used to produce vapor grown carbon fibers (VGCFs) as depicted. As shown in Figure 4, there are two methods to synthesize VACNFs. First, the method based on “seeded catalyst on a flat plate” and second, the “floating catalyst method”. The first technique uses the catalysts seeded on a substrate within the reactor, while in another method, the catalyst is deposited on a selected substrate as a film by sputtering or evaporation techniques.

Firstly, the commonest way used thin films of Ni, Fe, or Co metallic catalyst [93, 94] or their alloys [95], Cu–Ni composition [96], nickel-copper-aluminum takovite [97], mixture of  $\text{Ni}(\text{OH})_2$ – $\text{Mg}(\text{OH})_2$  [98], ferrocene [99], copper titrate [100], and cobaltocene [101] as catalyst precursors. Moreover, a buffer-layer such as Ti was often used as a barrier to diffuse and promote particle formation. Secondly, it is a “pretreatment step”; Carrier gas such as ammonia ( $\text{NH}_3$ )

or hydrogen ( $\text{H}_2$ ) is introduced as the chamber to a few Torr, and the sample is heated to the desired temperature, usually 500–700°C. Plasma is then initiated for several seconds to minutes. Lastly, there is the carbon source gas (such as  $\text{C}_2\text{H}_6$  [102],  $\text{C}_2\text{H}_5\text{OC}_2\text{H}_5$  [103],  $\text{C}_2\text{H}_2$  [104],  $\text{C}_{60}$  [105]) in the plasma, which immediately initiates CNF growth. The later way uses the catalysts and the carbon source gas floating in the reactor space at the same time.

This paper focuses mainly on the PECVD technique from the aspects of metal catalyst, carbon source, processing method, and other techniques by virtue of the classical examples and the recent reports. Pham-Huu et al. [102] showed that large-scale and uniform carbon nanofibers with a diameter of about 50 nm can be synthesized using the catalytic decomposition of a mixture of ethane and hydrogen over a nickel catalyst decorating carbon nanotubes at 550–650°C. The attained CNFs need not subsequent purification, due to the use of carbon nanotubes as support, the high nanofiber yields, and the purity reached. Zou et al. [103] reported that CNFs were prepared by cocatalyst Zn and Fe powders deoxidation process using  $\text{C}_2\text{H}_5\text{OC}_2\text{H}_5$  as starting material at 650°C. The obtained nanofibers possessed diameters about 80 nm, lengths ranging from several micrometers to tens of micrometers, low graphitic crystalline, and good electrochemical properties. The cocatalyst synergetic effect is valuable in controlling the resultant nanofiber diameter for synthesis and the study of other carbonous materials. Yu et al. [104] synthesized carbon nanofibers using the thermal decomposition of acetylene by a copper nanocatalyst derived from cupric nitrate trihydrate at a low temperature of 260°C. The copper nanoparticle size has a considerable effect on the morphology of carbon nanofibers. Helical carbon nanofibers and straight carbon nanofibers laid on catalyst copper nanoparticles with a grain size less than 50 nm or within 50–200 nm. The present study further assumes that it is possible to control the diameter of CNFs by controlling the size of the catalyst particle. Zhang et al. [105] successfully prepared carbon nanofibers with a diameter of about 100 ~ 500 nm using  $\text{C}_{60}$ , graphite-carbon, and boron powders via the ultrasonic spray pyrolysis method of ethanol without using metal catalysts. The experiment indicates that the clusters composed of carbon- and boron-related materials act as nucleating sites for CNFs formation. Kimura et al. [106] synthesized CNFs (lengths of 1–20  $\mu\text{m}$ , uniform diameters of 20–100 nm) with high electrical conductivity by ion beam irradiation of decacyclene whiskers at ambient temperature. This novel method of preparation without catalyst is advantageous for aligning the fibers as desired. The mechanisms of CNFs formation were shown in detail and found that CNFs were not grown from the film but were generated by the conversion of pre-existing whiskers of decacyclene on the film. Matsumoto et al. [107] described that carbon nanofibers were obtained using a hot filament-assisted sputtering system with pure argon gas as the sputtering gas, with a tungsten hot filament as the thermal electron emission source. Guláš et al. [108] observed that CNFs and related structures with outer diameters 20–60 nm were prepared by combination of aerosol synthesis and plasma-enhanced catalytic chemical vapor deposition with

alcohol as carbon precursor catalytic CVD (PE CCVD).  $\text{Al}_2\text{O}_3$  and Si coated Fe and Ni were deposited as catalysts. Ethanol and isopropyl alcohol vapors were used as a carbon source. HCGD is used as a gas activation process without any specific heating of the substrate that plays an important role in gas decomposition and activation for CNFs growth. On the other hand, Ren et al. [109] reported a simple synthesis method of carbon nanofibers using combustion of ethyl alcohol. Copper plate was employed as substrate, iron nitrate, and iron chloride as catalyst precursor and ethanol as carbon source. Carbon nanofibers with diameters of 10–100 nm were produced in bulk by the floating catalyst method by Ci et al. [110]. Several experiments showed the growth of carbon fibers depending on the reactor conditions and experimental parameters, so it was easily controlled to synthesize the large scale of carbon nanofibers. Mori and Suzuki [111] demonstrated that vertically aligned free-standing CNFs can be synthesized at low temperature as low as  $90^\circ\text{C}$  by plasma-enhanced chemical vapor deposition at low temperature CO/Ar DC plasma. The low-temperature synthesis of carbon nanofibers was demonstrated that the addition of a small amount of  $\text{O}_2$  was favorable for the synthesis of CNFs, because it suppresses the deposition of amorphous carbon.

**2.2.2. Carbon Nanotubes.** Carbon nanotubes are rolled up into tubular structures by  $\text{sp}^2$ -bonded graphite sheets with nanometer diameter and large length ratio. The nanotubes may consist of two different types of carbon nanotubes. Namely, singlewall nanotubes (SWNTs) made of single layers of graphene cylinders with typical diameter of the order of 1.4 nm and the multiwall nanotubes (MWNTs) made of 4–24 concentric cylinders of graphene layers with adjacent shells separation of 0.34 nm and a diameter typically of the order 10–20 nm. Nowadays, carbon nanotubes are still mainly synthesized by the arc-discharge, laser-ablation (vaporization), and chemical vapor decomposition (CVD) method.

The MWNTs were first observed which deposited on the negative electrode during the direct current arc-discharge of two graphite electrodes for preparation of fullerenes in an argon-filled vessel by Iijima in 1991 [112]. Large-scale and high-quality MWNTs were achieved by arc-discharge technique first by Ebbesen and Ajayan in 1992 [113]. In 1993, singlewall carbon nanotubes (SWNTs) are almost simultaneously obtained by arc-discharge and catalyst-assisted arc-discharge by Iijima and Ichihashi [114] and Bethune et al. [115]. Iijima and Ichihashi [114] used arc-discharge chamber that installed two vertical thin electrodes with a small piece of iron filled with a gas mixture of methane and argon. On the other hand, Bethune et al. [115] reported that SWNTs can be obtained by arc-discharge of anode thin electrodes with bored holes powdered metal catalysts (Fe, Ni or Co) incorporated into anode. As the arc method, metal catalysts were needed for the growth of SWNTs in contrary to MWNTs. Journet et al. [116] obtained large quantities of SWNTs by arc-discharge using a carbon anode with yttrium and nickel as catalysts. In 1996, high-yield SWNTs were produced using laser-ablation method of graphite with Ni and Co catalysts at

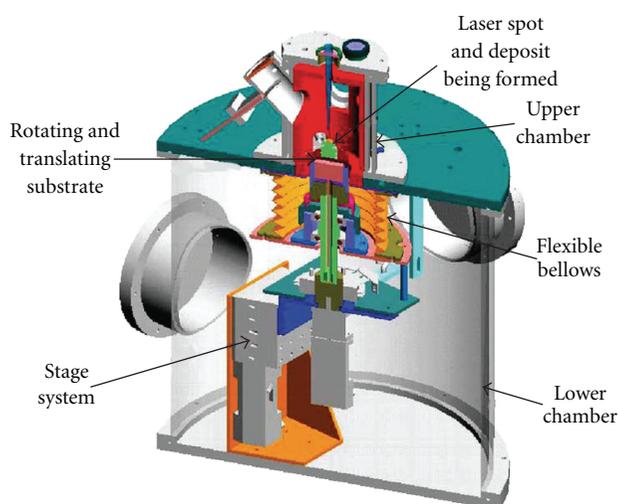


FIGURE 5: The schematic diagram of CNTs fabrication using the laser CVD reactor [129].

$1200^\circ\text{C}$  by Thess et al. [117]. The techniques based on arc-discharge and laser-ablation both have the advantage of high yield but also face the problem of high temperature needed and impurity. The chemical vapor deposition has been used successfully for producing carbon fiber and filament since the 1960s. Yacaman et al. [118] first produced the MWNTs by chemical vapor decomposition method in 1993. Flahaut et al. [119] made bulk amounts of SWNTs using CVD of methane catalyzed on mixed oxide spinels. When using CVD for synthesized CNTs, catalysts brought high yield because the increased metal-supporting interactions were propitious to the growth of carbon nanotube, which also caused impurity and aggregation of the metal nanoparticles. A diagram of CNTs fabrication by the method of CVD is schematically shown in Figure 5. Vast amount of reviews on carbon nanotubes have been discussed in the recent literature in detail, including the synthesis and growth mechanisms of CNT [120–129]; therefore, we described only three main productions in brief.

### 2.3. Two-Dimensional Carbon Nanomaterials (2-DCNs)

**2.3.1. Graphene.** Graphene, one-atom-thick planar sheet of  $\text{sp}^2$ -bonded carbon atoms, is arranged densely in a two-dimensional hexagonal honeycomb crystal lattice. There are three extremely strong  $\sigma$  bonds in-plane result in the mechanical stability of the carbon sheet,  $\pi$  orbitals perpendicular to the plane interactions between graphene and a substrate or between graphene layers are responsible for the electron conduction. It is the basic building block of (0-D) fullerenes, (1-D) carbon nanotubes, and (3-D) graphite.

In a large scientific community, more allotropes of carbon have been reported in succession. Diamond and graphite have been known for centuries, and the recently discovered fullerenes and nanotubes also have been studied in the last two decades. For a long time, graphene was

only considered as theoretical concept. Until 2004, [130] a physicists group led by Andre Geim and Kostya Novoselov from Manchester University, UK used mechanical exfoliation approach to obtain graphene. The discovery of isolated graphene monolayer has attracted wide attention to investigate the properties of this new yet ancient two-dimensional carbon nanomaterial due to its exceptional electronic and mechanical properties. More and more simple methods were searched for the growth of graphene. Several typical methods have been developed and reviewed as follows.

*Mechanical Exfoliation.* As mentioned above, [130] graphene flakes were first produced by continuously cleaving a bulk graphite crystal with a common adhesive tape and then transferred the thinned down graphite onto a cleaned oxidized silicon wafer substrate with visible color. The technique started with three-dimensional graphite and extracted a single sheet (a monolayer of atoms) called mechanical exfoliation or micromechanical cleavage.

Until now, mechanical exfoliation of graphite is still the best method to provide a small amount of high-quality samples for the study of a variety of graphene properties. Furthermore, the venerable technique has been used easily to obtain large size (up to  $100\ \mu\text{m}$ ), high-quality, two-dimensional graphene crystallites, which immediately brought enormous experimental researches [131–134]. Meanwhile, modified techniques are needed to provide a high yield of graphene for industrial production.

*Epitaxial Growth.* Recently, graphene was obtained by the epitaxial growth of graphene layers on metal carbides using thermal desorption of metal atoms from the carbides surface, or directly on metal surfaces by chemical vapor deposition (CVD). The typical carbide is SiC; silicon carbide heated to very high temperatures leads to evaporation of Si and the reformation of graphite; the control of sublimation results in a very thin graphene coatings over the entire surface of SiC wafers, which initially showed more performances than devices made from exfoliated graphene. So far, all of known synthesis approaches, however, are required in specialized laboratories for graphene sheets whose electronic properties are often altered by interactions with substrate materials. The development of graphene required an economical fabrication method compatible with mass production. The latest modified method was demonstrated by Aristov et al. [135]. Based on their work, for the first time, graphene was synthesized commercially on available cubic  $\beta$ -SiC/Si substrates, which was a simple and cheap procedure to obtain industrial mass production graphene, which meets the need of technological application. Moreover, many other types of carbide have been exploited to produce supported graphene, such as TiC (111), TiC (410), and TaC (111). It is well proved that metal surfaces can efficiently catalyze decomposition of hydrocarbons into graphitic materials to support growth of graphene on metallic surfaces by CVD. The advantage of epitaxial growth is large-scale area, but it is difficult to control morphology, adsorption energy, and high-temperature process.

*Chemical Exfoliation.* The theory of chemical exfoliation is to insert reactants in the interlayer space for weakening the van der Waals cohesion. At first, the graphite flakes are forced upon oxidative intercalation of potassium chlorate in concentrated sulphuric and nitric acid, received carbon sheets with hydroxyl and carboxyl moieties. The suspension is known as graphite oxide (GO). The GO is highly dispersible in water, and it can be easily deposited onto SiO<sub>2</sub> substrates. The precipitate of GO is sonicated to form separated graphene oxide sheet, then another reduction, and finally graphene sheet is formed. When KClO<sub>3</sub> is used, it generates a lot of chlorine dioxide gas and emits a great deal of heat, so the mixture is highly hazardous [136]. In 1958, Hummers and Offeman [136] reported a modified method which was much faster and safer. Based on the technique introduced by them, graphite is dispersed into a mixture of concentrated sulfuric acid, sodium nitrate, and potassium permanganate in contrast to KClO<sub>3</sub> [137]. Meanwhile, it must use H<sub>2</sub>O<sub>2</sub> to eliminate the MnO<sub>2</sub> generated from KMnO<sub>4</sub> time after time. This process faced a similar situation when it used m-CPBA [138] as an oxidant. Chandra et al. [139] also reported a novel synthetic route using oxidation acidified dichromate, to get high quality and stable aqueous dispersed graphene sheets. After GOs were deposited, chemical reduction of GO was accompanied by the elimination of epoxy and carboxyl groups using different reductants such as hydrazine [140], dimethylhydrazine [141], hydroquinone [142], and NaBH<sub>4</sub> [143], under alkaline conditions [144] or with thermal methods [145]. Because the reductants are usually hazardous, there are lots of interests on green routes to speed deoxygenation of graphene oxide. Wakeland et al. [146] introduced an approach to synthesize graphene from GO using urea as expansion reducing agent heated in an inert gas environment (N<sub>2</sub>) for a very short time to a moderate temperature (600°C). Chen et al. [147] successfully achieved thermal reduction of graphene oxide (GO) to graphene with the assistance of microwaves in a mixed solution of N, N-dimethylacetamide, and water (DMAc/H<sub>2</sub>O). The reduction of GO can be accomplished rapidly and mildly. This method is rapid, not requiring any solvents or stabilizers, inexpensive, and easy to scale up.

2.3.2. *Carbon Nanowalls.* Carbon nanowalls (CNWs) consist of vertical aligned graphene sheets standing on the substrates, form two-dimensional wall structure with large surface areas and sharp edges. The thickness of CNWs ranges from a few nm to a few tens nm. So far, research groups have explored different synthesis methods of CNWs based on plasma-enhanced chemical vapor deposition techniques. The main approaches are as follows.

- (1) Microwave plasma-enhanced chemical vapor deposition (MWPECVD).
- (2) Radio-frequency plasma-enhanced chemical vapor deposition (RFPECVD) (RF inductively coupled plasma (ICP) and RF capacitively coupled plasma (CCP)).
- (3) Hot-wire chemical vapor deposition (HWCVD).

- (4) Electron beam excited plasma-enhanced chemical vapor deposition (EBEPECVD).

For the first time, carbon nanowalls were accidentally discovered during the growth of carbon nanotubes by Wu et al. [148] using MWPECVD. In the experiment, the NiFe-catalyzed substrate (Si, SiO<sub>2</sub>/Si, sapphire) was preheated to about 650–700°C in hydrogen plasma; the mixtures of CH<sub>4</sub> and H<sub>2</sub> were utilized as flow gases. The nanowalls were monitored using SEM in different growth stages. The well-controlled MWPECVD synthesis process induced further studies to search more flexible control of the growth of CNWs, which aided to understand the mechanisms of CNWs growth and solving unwanted byproduct owing to the use of metal catalyst particles.

Recently, some groups have prepared CNWs without catalysts, using RFPECVD, assisted by a hydrogen atom injection. Shiji et al. [149, 150] synthesized carbon nanowalls on a Si substrate without catalysts using capacitively coupled RFPECVD by H atom injection. The grown samples employed fluorocarbon/hydrogen mixtures, used C<sub>2</sub>F<sub>6</sub>, CF<sub>4</sub>, CH<sub>4</sub>, and CHF<sub>3</sub> as the carbon source gas, and heated a substrate temperature of 500°C. The SEM images monitored the grow of carbon nanowalls using different system and the vacuum ultraviolet absorption spectroscopy (VUVAS) measured H atom density in the plasma in order to discuss the growth mechanism of carbon nanowalls. The experiment demonstrated that the structure and growth rate of carbon nanowalls depended on the types of carbon source gases and H atoms and played an important role for the formation of carbon nanowalls.

Considering the practical applications of carbon nanowalls, large scales of CNWs using MWPECVD and RFPECVD are difficult to provide. A promising method such as HWCVD enables to access the large scales easily compared with the above two methods. Moreover, HWCVD/CAT-CVD have the additional advantage of a high-hydrogen radical density.

Itoh et al. [151] reported that CNW films have been successfully prepared using only CH<sub>4</sub> by HWCVD. In the growth of CNWs, a substrate heated over 500°C would be needed. The hydrogen H<sub>2</sub> pressure is of 133 Pa by radical treatment. The structure of the CNWs has been studied by scanning electron microscope (SEM) and Raman spectroscopy. There are two reports about CNWs synthesis using CH<sub>4</sub> with different hydrogen dilution [152] and without hydrogen dilution [151] by HWCVD. Furthermore, Mori et al. [153] studied fabrication of vertically aligned and definite CNWs by an EBEPECVD employing a mixture of CH<sub>4</sub> and H<sub>2</sub> at relatively low temperature of 570°C. Figure 6 shows a schematic diagram of CNWs fabrication using EBEPECVD. An EBEP is a high-density plasma directly obtained by a high-current and low-energy electron beam. CNWs were characterized using SEM, TEM, and MRS [153].

According to the recent progress report [154], carbon nanowalls with large-surface areas and sharp edges were obtained on nickel-coated oxidized silicon substrates by PECVD in a radiofrequency Ar plasma beam injected with acetylene at temperature of 600°C. The formation of carbon

nanowalls was proved by SEM, TEM, ED, and Raman spectra. The mentioned study showed that the shape, surface distribution, and size of the nanostructures depended on the nature of the active gas and on the gas mass flow ratio in the Ar/H<sub>2</sub>/C<sub>2</sub>H<sub>2</sub> mixture.

Different from previous reports, various growth mechanisms and deposition parameters (catalyst, the effect of localized plasma) of CNWs were explored on a flat substrate, where existed a different growth mechanism. Non-surface-bound (freestanding) growth of CNWs by the method of MWPECVD without using the metal catalyst under a gas mixture of C<sub>2</sub>H<sub>2</sub> and NH<sub>3</sub> was developed by Chuang et al. [155]. A growth stage was used to facilitate energy-intensive, localized plasma for the growth of carbon nanowalls. Owing to the freestanding nature of their growth and the absence of catalyst, the synthesis process easily obtains a large scale of carbon nanowalls without impurity by metal catalyst particles and processed for various applications. There is a similar report [156] of freestanding graphite sheets with thickness less than 1 nm, in which “carbon nanosheets” were synthesized on a variety of substrates by RFPECVD without any catalyst or special substrate treatment. Further studies will be required to understand the formation of non-surface-bound (freestanding) carbon nanowall materials.

Sato et al. [157] investigated the density peak in argon plasma and found that the helicon wave was effectively excited and produced the plasma antenna. For the first time, the helicon-wave discharge was demonstrated to be used as PECVD source for the growth of CNWs. Using the helicon-wave reactive plasma as a precursor source for plasma-enhanced chemical vapor deposition, well-aligned carbon nanowalls are found to be formed even in a very low gas pressure of 0.7 Pa. Table 1 briefs the main synthesis methods of different low-dimensional carbon nanomaterials as described in this section. Further development in this field is expected on their rich physics and potential applications.

### 3. Properties and Applications

There are several various allotropes of carbon such as graphite, diamond, and amorphous carbon. Therefore, the physical and mechanical properties of carbon strongly depended on the allotropic forms of carbon. As an example for the mechanical property of hardness, diamond is known as one of the hardest materials, while graphite is soft enough to be used for making pencils. About the property of color, diamond is considered transparent while graphite is an opaque material and black. As another example, while graphite is a good conductor, diamond just demonstrates a low electrical conductivity. On the other hand, diamond is normally known as a highly thermal conductive, while graphite is considered as the most thermodynamically stable material.

**3.1. Fullerene.** As described in Section 2.1.1, fullerene belongs to zero-dimensional carbon nanomaterials. Applications of fullerene include the applications in medicine such as their aid to produce the specific antibiotics and drugs for

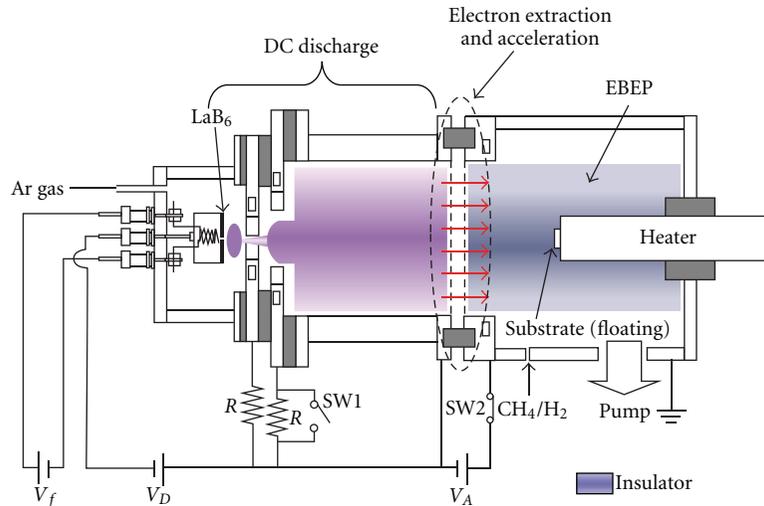


FIGURE 6: The schematic diagram of CNWs fabrication using EBEPECVD [153].

TABLE 1: Comparison of the main synthesis methods of different low-dimensional carbon nanomaterials.

LDC-nanomaterial	Synthesis methods
Fullerene	Laser vaporization, Arc discharge, Combustion, Microwave plasma Pyrolysis, Flat flames
CEMNPs	Arc-discharge, high-temperature heat treatment, mechanical milling, Co-carbonization, Pulse-laser irradiation
ND	MPCVD, Hot-filament, CVD, Pulse laser ablation, Electro irradiation, High-energy X-ray diffraction
OLCs	Arc-discharge, High-electron irradiation, CVD, RFP Thermal annealing of diamond nano-particle
CNFs	Traditional vapor growth, Catalytic combustion, PECVD, Hot filament-assisted sputtering, Ultrasonic spray pyrolysis, Ion beam
CNTs	Arc-discharge, Laser-ablation, CVD, Other methods
Graphene	Mechanical exfoliation, Epitaxial growth, Chemical exfoliation
CNWs	MWPECVD, RFPECVD, HWCVD, EBEPECVD

certain cancers particularly melanoma. On the other hand, due to their specific properties, fullerenes have had great applications in the field of superconductivity.

In 1991, Haddon et al. [158] reported the preparation of alkali-metal-doped films of C<sub>60</sub> and C<sub>70</sub> which have electrical conductivities at room temperature that are comparable to those attained by n-type-doped polyacetylene. They observed the highest conductivities in the doped films equal to 4 Scm<sup>-1</sup> (Cs/C<sub>60</sub>), 100 (Rb/C<sub>60</sub>), 500 (K/C<sub>60</sub>), 20 (Na/C<sub>60</sub>), 10 (Li/C<sub>60</sub>), and 2 (K/C<sub>70</sub>) [152]. In the same year, Hebard et al. [159] reported that potassium-doped C<sub>60</sub> becomes superconducting at 18 K [159]. As the researchers stated in their paper [159], the observed superconductivity at 18 K was the highest yet observed for a molecular superconductor. Furthermore, the superconductivity in fullerene doped with various other alkali metals was reported by some other researchers [160, 161]. Subsequently, Zhou et al. in 1992 [162] and Brown et al. in 1999 [163] showed that the superconducting transition temperature in alkaline-metal-doped fullerene was elevated while the unit-cell volume *V* was increased. In 2008, Ganin et al. [164] reported the bulk superconductivity at 38 K in a molecular system. The system introduced by the researchers was bulk Cs<sub>3</sub>C<sub>60</sub> as a key material in this family [164], and as the authors explained,

cesium-doped fullerene is an important material in the same family [164].

On the other hand, fullerenes are almost stable chemically, however, not completely uncreative. In the view of the solubility, apart from some fullerene structures which are not soluble due to the presence of a small band gap in their structure between the ground and excited states, they are generally soluble in various solvents. In fact, fullerenes are the only carbon allotropes which can be dissolved in common solvents at room temperature. The mentioned common solvents can be aromatics such as toluene and carbon disulfide.

**3.2. Nanodiamond Properties.** The physical properties of nanodiamonds (NDs) are briefed in Table 2 [165]. Due to the unique structure of nanodiamonds, they are known as the nanomaterials with significant properties. In fact, 30 percent of nanodiamond atoms are located on the surface of their structure. The maximum size of the single grains of nanodiamonds is 10 nm, while the average particle size is obtained between 4 and 6 nm [50, 166].

In 2000, Iakoubovskii et al. [50] applied the method of X-ray diffraction (XRD) to study the characterization of the structure and defects in detonation synthesis and

TABLE 2: Physical properties of nanodiamond [165].

Structure	Cubic ( $a = 0.3573$ nm)
Particle size	4.5 nm
Number of C atoms	ca. 10,000
Molar mass	120,000 g
Specific surface area	300 m <sup>2</sup> /g
Graphitization temperature	1100° C
Zeta potential	-78.44 mV
Refractive index	2.55

ultradisperse diamond (UDD). In 2008, Iakoubovskii et al. have applied the method based on high-resolution transmission electron microscopy (HRTEM) [166]. They have showed that the surface area of nanodiamond is relatively large, and the size of diamond grains is distributed around 5 nm. According to a FTIR study of the adsorption of water on ultradispersed diamond powder surface by Ji et al. [167], the surface of detonation nanodiamonds can naturally adsorb the water and hydrocarbon molecules from the environment atmosphere [167]. As the result of the research work by Iakoubovskii et al. using HRTEM, although the detonation nanodiamond grains were mostly cubic lattice, yet they were not perfect structures. According to the results of HRTEM, the main defects are shown as the multiple twins [166].

Applications of nanodiamonds cover a wide range, mainly including the commercial products such as the polishing products, additives to engine oils, reinforcing fillers for plastics and rubbers, dry lubricants for metal industry, and their applications in galvanic electrolytes. On the other hand, the recent researches on application of nanodiamonds in medical science particularly in chemotherapy drugs production are being performed.

**3.3. Carbon Nanotubes.** Due to the amazing structure and properties of CNTs, these materials have found great applications in very wide areas of science and technology including nanotechnology, electronics, optics, materials science, and architecture. In the view of the structural applications of CNTs, these applications cover a wide ranges of industrials such as clothes, sports equipment such as stronger and lighter tennis rackets, bike, various kinds of balls, combat jacket like combat jackets, concrete as the increase of the tensile strength, polyethylene, the possibility of the space elevator, synthetic muscles in medical science and sports, high tensile strength fibers, applications in the build of bridges, ultrahigh-speed flywheels, and applications in fire protection.

In electromagnetic field, the applications of CNTs are briefed in respect of their uses in chemical nanowires, conductive films, electric motor brushes, magnets, optical ignition, their applications to produce light bulb filament (as an alternative for tungsten filaments), the applications related to their fine superconductivity properties, in display screens such as field emission displays (FEDs), the

applications in transistor industrial, and the electromagnetic antenna.

Other applications of CNTs are briefed in respect of their chemical applications, including air pollution filters, biotech containers, hydrogen storage, water filtration, and the mechanical applications such as using them as the faster oscillators, nanotube membrane, slick surface, carbon nanotube actuators, infrared detector, radiometric standard, and their application as the thermal radiation for space satellites.

This is worth to know that the wide range of applications of CNTs mentioned above is due to their unique mechanical, electrical, thermal, and optical properties which are briefly introduced in this section.

**3.3.1. Strength.** Carbon nanotubes (CNTs) are the strongest materials yet discovered, which is due to the covalent sp<sup>2</sup> bonds between the individual carbon atoms. In 2000, Yu et al. [168] tested a multiwalled carbon nanotube (MWCNT) to access the tensile strength up to 63 GPa. The specific strength is defined as the material's strength (force per unit area at breaking point) divided by its density. Considering the low density of CNTs (1.3 to 1.4 g cm<sup>-3</sup>), the specific strength of CNTs is obtained up to  $48 \times 10^3$  kN m kg<sup>-1</sup>. This value is dramatically the best of the known materials.

To compare the strength property of various CNTs to some other strong materials such as stainless steel and kevlar, the tensile strength and Young's modulus of these materials have been briefed in Table 3 [168–175].

The first row of Table 3 shows the data for SWNT ropes based on the “experimental” measurements by Yu et al. using the microscopy method based on AFM [171, 173]. Yu et al. [173] measured the mechanical responses of 15 SWCNT ropes under the tensile load [173]. As they have stated in their paper [173], they considered all SWNT samples as (10, 10) nanotubes, with the measured diameter of 1.36 nm [173]. According to their experiments, as it is seen in Table 3, the average breaking strength value varies from 13 to 52 GPa [173]. Furthermore, in the same evaluation, Yu et al. measured the average Young's modulus values of the SWNT rope samples equal to 1002 GP (from 320 to 1470 GPa or about 1 TP in Table 3). In the same study, the failure strains occur at 16%. In addition, Yu et al. reported the similar data for 19 samples of MWNTs. Based on their report [173], the strength and Young's modulus values for MWNT samples vary from 11 to 63 GPa and 270 to 950 GPa, respectively (Table 3). In addition, Demczyk et al. [174] observed the force required to break the samples of MCNTs directly using transition electron microscopy. Based on the conducted pulling and bending tests on individual MWNTs, the mentioned researchers have measured the tensile strength and elastic modulus of MCNTs equal to 150 GPa and 800 GPa, respectively [174].

On the other hand, the second and third rows of Table 3 show the related data as the “predicted” strength tensile values based on the analytical simulation models. Various “analytical” simulations and models have been recently applied to calculate the tensile and Young's modulus values for CNTs. An interesting model based on a molecular

TABLE 3: Comparison of the mechanical properties of various nanotubes [168–175].

Material	Tensile strength (GPa)	Young's modulus (GPa)	Failure strain (%)
SWNT	13 to 53 (measured)	1002 (~1 TPa) (measured)	16
Armchair SWNT	126.2 (predicted)	940 (predicted)	23.1
Zigzag SWNT	94.5 (predicted)	940 (predicted)	15.6–17.5
Chiral SWNT	—	920	—
MWNT	11 to 150	270 to 950	—
Stainless steel	0.38 to 1.55	186 to 214	15 to 50
Kevlar	3.6 to 3.8	60 to 180	~2

mechanics by Xiao et al. [175] predicts the maximum values of 126.2 GP and 94 GP for Armchair SWNT and Zigzag SWNT, respectively. As given in Table 3, the predicted failure strains in the study by Xiao et al. are 23.1% and within 15.6–17.5% for armchair nanotubes and zigzag nanotubes, respectively [175].

Generally, as it is seen from Table 3, both the Young modulus and tensile strength of various carbon nanotubes are significantly more elevated than those for stainless steel and kevlar. It seems that CNTs are tended to a permanent deformation under a strong tensile strain. Furthermore, due to the hollow structure and high aspect ratio of CNTs, it seems that their strength is limited under compression or bending stress. On the other hand, as it can be seen from the simple geometry of CNT, carbon nanotubes should be softer in the radial direction than along the tube axis. In 1993, Ruoff et al. studied the Radial deformation of carbon nanotubes by van der Waals forces using transmission electron microscopy (TEM) [176]. According to their suggestion, two near nanotubes can be deformed even by van der Waals forces. Furthermore, according to the studies by Yu et al. in 2000 on radial deformability of individual carbon nanotubes under controlled indentation force (Figure 7) [177] and by Palaci et al. in 2005 [178] on radial elasticity of multiwalled carbon nanotubes, as can be seen in Figure 8, carbon nanotubes are rather soft in their radial direction.

**3.3.2. Hardness.** In 2002, Popov et al. [179] reported the synthesis of a super-hard phase (SP) composed of single-wall carbon nanotubes, measured by a nanoindenter within 62–152 GPa. The synthesis treatment was performed by compressing SWNTs to above 24 GPa at room temperature. The authors considered the hardness of diamond and boron nitride samples (150 and 62 GPa, resp.) as mentioned in the references in their work [179]. The high mechanical properties of SP-SWNT, namely bulk modulus, were obtained as 462–546 GPa, surpassing the value of 420 GPa for diamond [179]. The authors have found the hardness of SP-SWNT in the range of 62 to 150 GPa and attributed SP-SWNT to the class of super-hard materials [179].

**3.3.3. Electrical Properties.** The electrical properties of a nanotube strongly depend on its structure due to the symmetrical and exceptionally electronic structure of graphene. For example, as shown in Figure 9, in a graphene crystal lattice,  $n$  and  $m$  are the number of unit vectors along

two directions in which the graphene sheet is wrapped to make nanotube, so that  $(n, m)$  is called the chiral vector. Zigzag nanotubes are formed by wrapping a graphene sheet whenever  $m = 0$ , and in the same way, the structure is called armchair nanotube if  $n = m$ ; Otherwise, they are called chiral nanotubes. In the view point of the electrical properties of CNTs, according to the model described by Lu et al. mentioned above [180], armchair nanotubes ( $n = m$ ) are metallic. On the other hand, if  $n$  is not equal to  $m$ , however,  $(n - m)$  is a multiple of 3, the nanotube is semiconducting with a very small band gap; otherwise, the nanotube is a moderate semiconductor [180].

According to the mentioned rule, the structure of  $(5, 0)$  SWNTs should be semiconductor; however, exceptionally according to the calculations, due to the curvature effects in small diameter carbon nanotubes, this structure is metallic [180]. On the other hand, zigzag and chiral SWNTs with small diameters that are expected to be metallic have actually a finite gap [180].

In general, as Zhou et al. [181] have mentioned, SWNTs show excellent electronic properties such as the carrier mobility about  $10^4 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  [182] which is higher than that of silicon. On the other hand, as Dai et al. have described [182, 183], CNTs can carry an electrical current density of about  $4 \times 10^9 \text{ A cm}^{-2}$ . Such a current density is three orders of magnitude higher than a typical metal, for example, Cu or Al [182].

Furthermore, in order to describe the electrical properties of multiwalled carbon nanotubes (MWNTs), it is worth to mention that, in 2006, Takesue et al. [184] have confirmed that the entirely end-banded MWNTs exhibit superconductivity with a transition temperature as high as 12 K which is approximately 30 times greater than that for ropes of SWNTs or for MWNTs with usually noninterconnected shells [184].

**3.3.4. Thermal Properties of CNTs.** As shown in Figure 10, the thermal conductivity of an SWNT at room temperature along its axis with the length  $L \approx 2.6 \mu\text{m}$  and the diameter  $d \approx 1.7 \text{ nm}$  was reported as  $k \approx 3500 \text{ W m}^{-1} \text{ K}^{-1}$  by Pop et al. [185]. In order to compare the reported value by Pop et al. [185] with a good metallic thermal conductor, Cu shows a thermal conductivity about  $385 \text{ W m}^{-1} \text{ K}^{-1}$ . Furthermore, as Thostenson et al. [186] have mentioned, SWNTs show the stability to the temperature up to  $2800^\circ \text{C}$  in vacuum and  $750^\circ \text{C}$  in air, while metallic wires in microchips are melted at  $600\text{--}1000^\circ \text{C}$  [186].

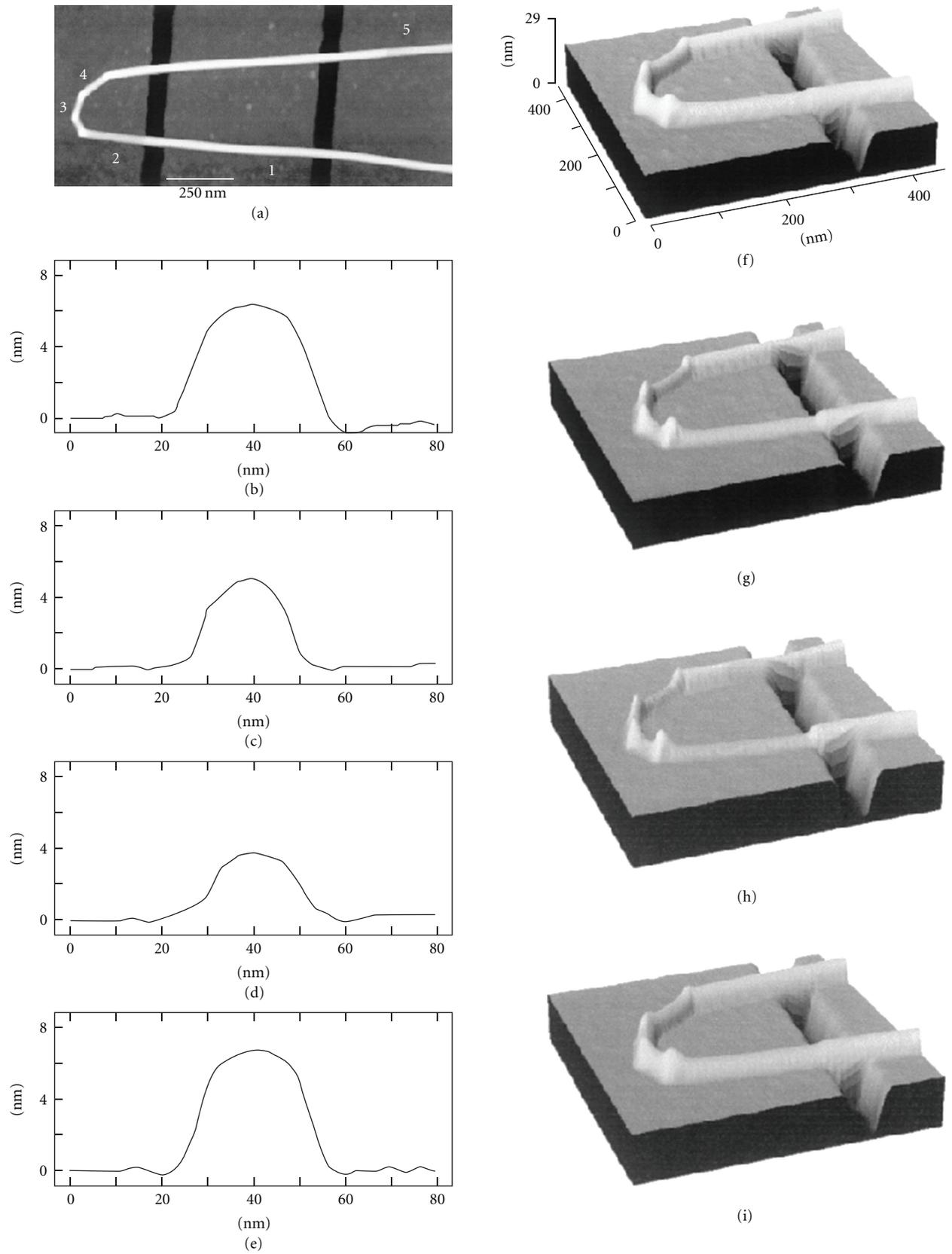


FIGURE 7: Deformability of a MWCNT deposited on an patterned silicon wafer as visualized with tapping-mode AFM operated far below mechanical resonance of a cantilever at different set points [177].

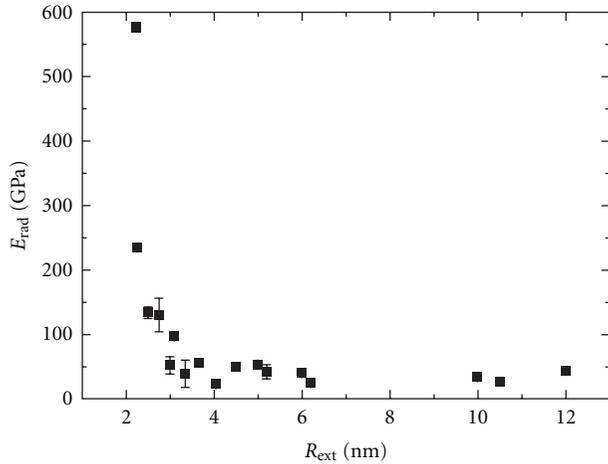


FIGURE 8: The radial Young modulus of CNTs as a function of  $R_{\text{ext}}$ , experimentally [178].

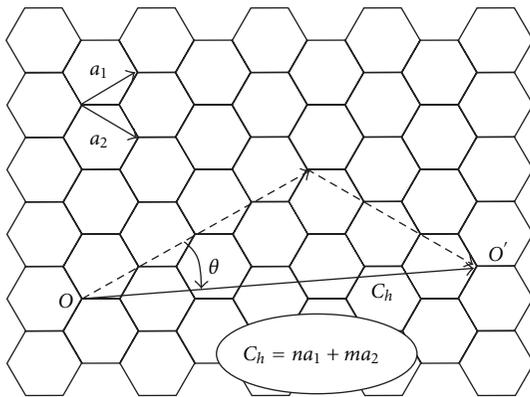


FIGURE 9: An  $(n, m)$  SWCNT: roll-up vector  $C_h = na_1 + ma_2$  and chiral angle  $\theta$  (for this special case,  $n = 5, m = 3$ ;  $a_1$  and  $a_2$  are the primitive vectors of a graphene sheet) [180].

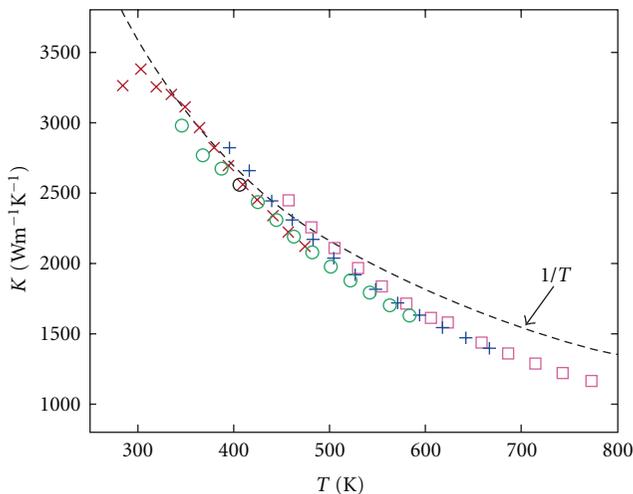


FIGURE 10: The relation between the extracted values of thermal conductivity and the average SWNT temperature [185].

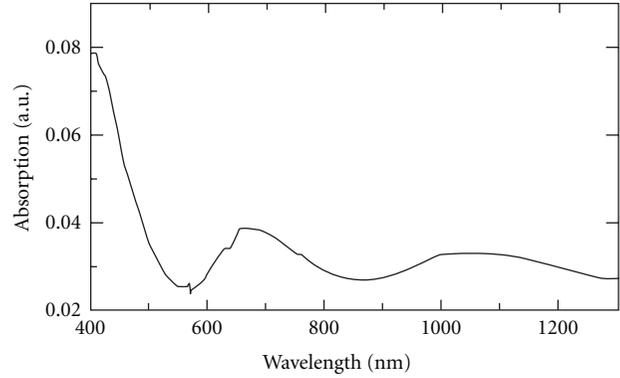


FIGURE 11: The adsorption spectrum of CNTs [172, 187].

3.3.5. *Optical Absorption.* The optical absorption in carbon nanotubes determined by Liu et al. [187] is shown in Figure 11. On the other hand, Margulis [188] has studied theoretically to indicate that zigzag SWNTs have positive low-frequency third-order susceptibilities. The Optical absorption in CNTs is occurred due to the electronic transitions from the levels  $v_2$  to  $c_2$  or  $v_1$  to  $c_1$ , and so forth [172, 189]. The mentioned transition is sharp, so it can be used to identify the type of nanotube. As it was shown by Itkis et al. [190], optical absorption is used to obtain the quality of the CNTs' powders [191].

On the other hand, according to an interesting study by Mizuno et al. in 2009 [191], SWNTs can have absorbances of 0.98–0.99 from the far-ultraviolet (200 nm) to far-infrared (200  $\mu\text{m}$ ) wavelengths. Therefore, compared to an ideal black body (with the absorbance of 1.0), a typical carbon nanotube is considered as a “practical black body” [191].

3.4. *Graphene.* In 2007, Meyer et al. [192] used transmission electron microscopy (TEM) to study the atomic structure of a single-layer graphene. As they reported, they have studied individual graphene sheets freely suspended on a micro-fabricated scaffold in vacuum or the air. The membranes were only one-atom thick, yet they still displayed long-range crystalline order [192]. According to their studies using TEM, the suspended graphene sheets are not perfectly flat. In other words, the suspended graphenes were observed as rippling of the flat sheet, with amplitude of about 1 nm [192]. As the authors explained, the atomically thin single-crystal membranes offered ample scope for fundamental research and new technologies, whereas the observed corrugations in the third dimension may provide subtle reasons for the stability of two-dimensional crystals [192].

3.4.1. *Mobility of Graphene and the Applications.* Graphene is known as a high electron mobility material at room temperature, so that the reported value is  $15,000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$  [193]. In 2005, Novoselov et al. [194] considered graphene as a condensed-matter system in which electron transport is essentially governed by Dirac's (relativistic) equation [194]. In 2008, Morozov et al. [195] studied temperature dependences of electron transport in graphene and showed

that the electron mobility higher than  $2 \times 10^5 \text{ cm}^2 \text{ V}^{-1} \text{ s}$  is achievable if extrinsic disorder is eliminated. In the same year, Chen et al. [196] studied the intrinsic and extrinsic performance limits of graphene devices on  $\text{SiO}_2$ . According to the studies mentioned above, it seems that the electron mobility in graphene should be almost independent of the temperature between 10 K and 100 K [194–196].

Due to the high mobility of graphene, this material is known as a promising nanomaterial particularly for those applications in which transistors need to switch extremely fast. Furthermore, the high mobility of graphene involves this material in the applications related to chemical and biochemical sensing.

On the other hand, the resistivity of the graphene sheets is  $10^{-6} \Omega \text{ cm}$  which is less than the resistivity of silver as known as the lowest resistivity at room temperature. Such a unique low resistivity and also the very low thickness of graphene have made this material to have a great role in many applications such as mechanical fields, electrical conducting, and transparent films which are necessarily applicable in the field of electronics such as producing touch screens and photovoltaic cells.

#### 4. Conclusion and the Future Outlooks

As described in this paper, the unique structure and properties of low-dimensional carbon nanomaterials as the advanced materials have led them to have a strong and important potential role in various scientific fields and engineering such as nanoscale electronic devices, field emission displays, diodes, transistors, sensors, composite polymers, artificial muscles, mechanical reinforcements, capacitors, and hydrogen storage. For example, carbon nanobuds are the recently produced materials from two previously known allotropes of carbon nanotubes and fullerenes. These fullerene-like “buds” have found the unique properties of both fullerenes and CNTs which have many applications as good field emitters as well as their role to improve the mechanical properties of composites. As another example, the application of CNTs to develop the biofuel products is being noticeably growing due to their strongly deferent properties comparing to the previous products.

The list of companies which are working on the new features of carbon nanomaterials can show the importance of the use of these key materials in industry as well as the scientific areas. Taken the examples of such companies and their products, we can mention the Eagle Windpower Ltd. (windmills with lightweight blades), Easton (on bicycle components), Xintek (on Nanotube-based cathodes, AFM probes, and X-ray tubes), Nanomix (sensors for detecting chemical vapors), Nano-Proprietary, Inc. (analyzing chemicals in liquid samples), Zyvex Performance Materials (epoxy resins strengthened with carbon nanotubes), Hyperion (Nanotube-based plastic mold compounds), Nano Lab (functionalized nanotubes and nanotube arrays), Nanoledge (Nanotube-based resins), Nanocs (Functionalized nanotubes), Nanocyl (nanotube-based epoxy resins), Amroy Europe Oy (Nanotube-based epoxy resins), Bayer Material

Science (Carbon nanotubes), Cheap Tubes (Carbon nanotubes), Catalytic Materials (Carbon nanotubes and graphite nanofibers), MER Corporation (Carbon nanotubes and buckyballs), NanoCarbLab (Carbon nanotubes), NanoAmor (Carbon nanotubes, carbon nanofibers, nanowires, and nanoparticles), Nanothinx (Carbon nanotubes), Rosseter Holdings (Carbon nanotubes), and many others.

As a matter of fact, due to the unique mechanical, optical, and electronic properties of carbon nanotubes, the publication statistics show that CNTs have succeeded to attract the main body of the authors' interest since 1991 up to now. However, as described in this paper, carbon nanomaterials are not limited to CNTs. Therefore, the future outlook of applications of these materials depends on the capability of the use of each one. As an interesting bioapplication example, nanodiamonds may be capable to be used for biolistic delivery in gene therapy, drug delivery, and vaccines as a solid support matrix. Furthermore, there is a strong possibility in near future to use nanodiamonds in the medical immunoassays as either the detection tag or the solid support matrix [197].

As the definition of the low-dimensional carbon nanomaterial, these materials also cover a wide range of carbon-related nanostructures such as nanodiamonds, fibers, cones, scrolls, whiskers, and graphite polyhedral crystals. In fact, there are expectable outlooks for the use of these materials in the fields of molecular electronics, sensing, nanoelectromechanic devices, field-emission displays, energy storage, and composite materials, as well as their growing applications in medical science, health, and daily life [198].

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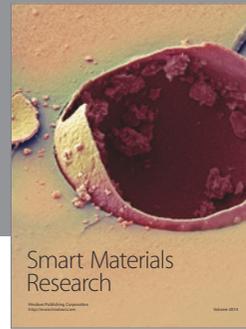
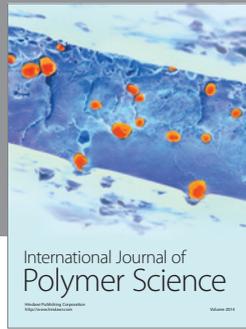
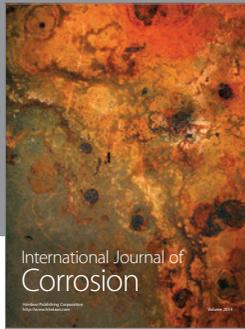
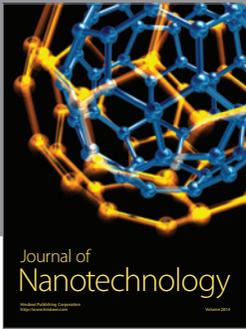
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