Chapter

Carbon Nanotubes

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Abstract

Carbon nanotubes (CNTs) are referred to as carbon nano-architecture allotropes, with wrapped graphene sheets forming a cylindrical structure. CNTs are either developed by metals or narrow-band semiconductors with rolling graphene sheets in various ways. Researchers have dedicated a great deal of attention to understanding the fascinating properties of CNTs over the years, and possess certain peculiar properties, such as a high degree of stiffness, a wide ratio of length to diameter, and remarkable toughness, and are employed in a number of applications. These properties can be enhanced by scheming the diameter, nature of walls, chirality, length of CNTs which is rolled up, and depending on the synthesis process. This chapter extensively covers the various properties of CNTs and how it influences to desired applications and also explains numerous methods of synthesis and processing of CNTs with advantages and some drawbacks.

Keywords: 2D materials, graphene oxide, carbon nanotube, arc discharge, laser ablation

1. Introduction

In recent decades, formation of nanowires and nanotubes has an attractive literature which emphasizes towards material growth. Amongst numerous materials containing organic and inorganic, nanotubes show versatile properties due to promising candidates such like carbon nanotubes, contributing a great part in potential applications relevant to disciplinary medicinal chemistry [1, 2]. Foundation of fullerenes [3] was extracted from carbon nanotubes (CNTs) that explore fabrication on a macroscopic level, thereby exhibiting continuous evolution [4]. The cylindrical shape of CNT is caused by rolling up of graphitic sheets; length is measured in micrometer scale while maximum diameter is taken as 100 nm. CNT also appears in bundle shape to form prominently complex nature structure [5]. Hexagon rings are in arranged form on which metallic nature or semiconducting behavior of CNT is evaluated. CNT belongs to the properties towards robust applications like fillers; bio-sensors are amongst nanotechnological pillars in exciting fields [6, 7]. However, some limitations such as insolubility and non-manipulation in solvents play role for creating hindrance to CNT use as solute in organic solvents as well as aqueous media. Dispersion of CNT may be carried out through sonication; however, precipitation is also occurred caused by the interruption of the process followed by the mechanism. Moreover, numerous studies also showed that CNT might react with a variety of chemical compounds [8–17].

Innovative nanodevices are greatly desired in research work and it may be met only by CNT best fabricating processing that is obtained by the synthesis of complex nature composites [18–20]. Furthermore, CNTs become highly reliable when chemical reactions are carried out to incorporate them in soluble activities into different systems such as organic or inorganic and biological accordingly. Thus, CNTs solubility approach in chemical reactions opens new routes for introducing promising materials [21, 22]. Unidirectional CNT structures may be prepared by modified approaches and their structural study is done by following group study containing three categories, first is that various chemical groups are incorporated on the surface of CNTs via covalent bonding, secondly non-covalent wrapping of functional groups and thirdly endohedral fulfillment of cavity. Many citations in this study have been appreciated due to which it is rapidly increasing by worth in literature, while this review presents a limited approach providing useful information in all citations followed in this study [23–26]. It has been systematically studied that CNTs may be prepared by employing synthesis methods containing arc discharge approach or chemical-vapor-deposition and laser-ablation technique [27, 28].

In arc discharge approach temperature is kept greater than 3000°C. This temperature is indispensable to evaporate carbon atoms to form a plasma state, in this way CNTs are shaped as single-walled as well as multi-walled structures. In this process, catalytic agent may or may not be involved during the formation of multi-walled carbon nanotubes (MWCNTs). However, inclusion of catalytic agent is mandatory to create individual single-walled carbon nanotubes (SWCNTs). The catalytic agents like Cobalt, Nickel, and Iron may be used as mandatory steps to complete the reactions reasonably [29–32]. In chemical-vapor-deposition (CVD) approach methane, ethylene, etc. are incorporated as hydrocarbon sources necessary to carry out reactions successfully. As far as laser-ablation approach is concerned, evaporation process of graphite occurs in a furnace at a temperature of 1200°C. Moreover, graphite appears as dominant material to produce species with converting ratio at maximum level. Moreover, biomaterial targets are achieved depending on degree of purity level, that is why macroscopic approach is carried out for the improved quality of carbon materials owing to achieve some characteristics like length and alignment [33]. It has been reported that MWCNTs were collected first time by Iijima (by employing arc-discharge approach), and this approach is too old that was adopted for carbon fibers synthesis [34, 35]. Subsequently, an in-situ emulsion of polymerization was presented by Khan et al. [36] in 2016 to synthesize carbon nanotubes (CNTs) in the form of composites, which was completed by employing a colloidal system to fabricate nanostructured brush.

2. Classification of carbon nanotubes

Nanotubes may be categorized into SWCNTs as well as MWCNTs (see **Figure 1**). A comparison between both SWCNT and MWCNT is demonstrated in **Table 1** [38, 41].

3. Structure and morphology

SWCNT comprised of carbon atoms from graphene sheet containing benzene rings in hexagonal shape as illustrated in **Figure 2a**. Cylindrical graphene sheets comprising honeycomb lattice are visualized in single-atomic graphitic-layer of crystalline nature, while MWCNT is in stacked form of graphene sheets that are rolled up into cylinders having same centers. The composition of nanotube

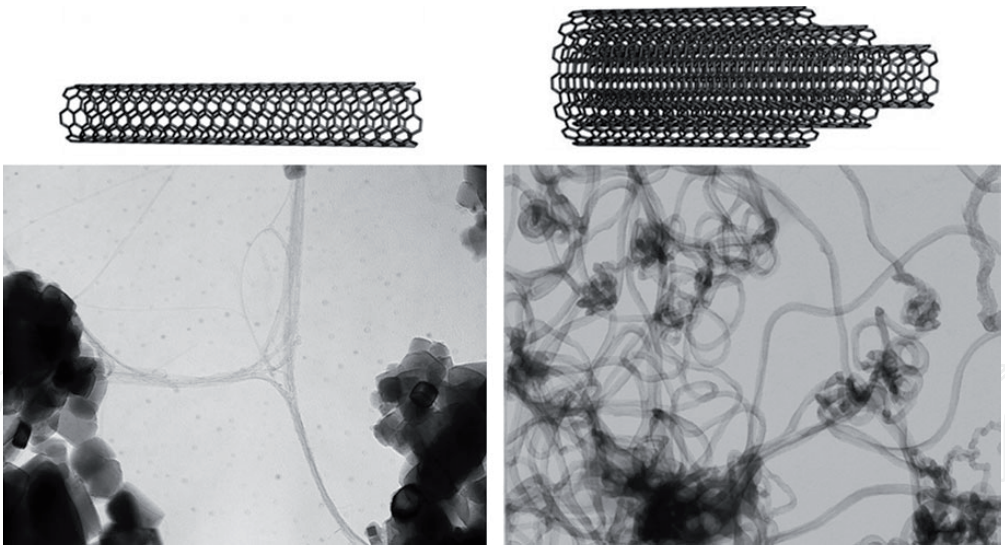

Figure 1.
Molecular representations of SWCNT (top left) and MWCNT (top right) with typical transmission electron micrographs below [37].

SWCNT	MWCNT
Single-layer of graphene.	Multiple layers of graphene
Catalyst is required for synthesis.	Can be produced without catalyst.
Bulk synthesis is difficult as it requires proper control over growth and atmospheric condition.	Bulk synthesis is easy.
Not fully dispersed, and form bundled bundled formation.	Homogeneously dispersed with no apparent structures.
Resistivity usually in the range of 10^{-4} – 10^{-3} Ω.m	Resistivity usually in the range of 1.8×10^{-5} – 6.1×10^{-5} Ω·m
Purity is poor. Typical SWCNT content in as-prepared samples by chemical vapor deposition (CVD) method is about 30–50 wt%. However high purity up to 80% has been reported by using arc discharge synthesis method.	Purity is high. Typical MWCNT content in as-prepared samples by CVD method is about 35–90 wt%.
A chance of defect is more during functionalization.	A chance of defect is less especially when synthesized by an arc-discharged method.
Characterization and evaluation are easy.	It has a very complex structure
It can be easily twisted and are more pliable.	It cannot be easily twisted.

Table 1.
Comparison between SWCNT and MWCNT [38–40].

molecules contains a million atoms having length of tens micrometers and diameters are comparable with 0.7 nm value [41]. SWCNTs containing 10 atoms often lie along the circumference of tube-like structure with one-atom-thick thickness. A length to diameter ratio of carbon nanotubes is measured about 1000 (large aspect ratio), giving rise to be considered as unidirectional structures [43]. MWCNTs structure is formed by various single-walled tubes that are stacked in concentric cylinders inside each other. The MWCNTs are identified as nanostructures showing the outer diameter is (15 nm or less) while structures having a diameter more than 15 nm are considered as nanofibers, not nanotubes. CNTs are different from carbon-fibers owing to not a single (molecule) yet strand layers sheets of graphitic nature [43–46].

Depending upon the two aforesaid basic structures, carbon nanotubes may be categorized into three varieties as an armchair, zigzag, and chiral carbon nanotubes.

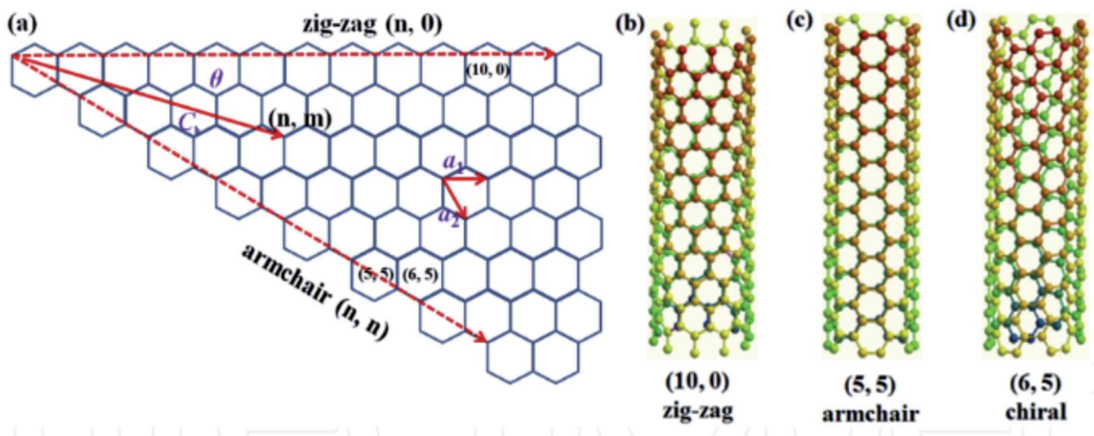

Figure 2.
(a) Unrolled single-layer graphene sheet showing the geometry of the SWCNT, (b-d) Examples of the three types of nanotube sidewall; zigzag, armchair, and chiral (A color version of this figure can be viewed online) [42].

The structure of a variety of carbon nanotubes depends on the formation of rolled up graphitic cylinders during synthesis process. The main focus is selection of rolling-axis relative to hexagonal graphitic network of sheets as well as radius of closing cylindrical network of nanotubes that are raised in various types of SWCNTs. In this structure, chiral vector contains *n* and *m* indices corresponding to two unit vectors directing along two-axis in graphene crystal lattice structure. In case of m = 0 zigzag-type nanotube, but when n = m the armchair nanotube is obtained while other configurations are attributed to chiral type nanotubes accordingly. In addition, SWCNTs with armchair structure, zigzag, and chiral structures have been illustrated in **Figure 2b–d**. Moreover, a further detailed structure may be visualized in literature reviews [7, 43, 47, 48].

4. Properties

Mechanical properties may drastically be raised, caused by the electrostatic forces between sp^2 carbon–carbon-bonds. Previously no material has been yet found to display the collective mechanical, electronic, and thermal properties up till now. Densities of materials have been observed below 1.3 g/cm^3 value (one-sixth stainless steel). Young's moduli measured material stiffness that was greater than 1 TPa and is considered approximately 5x higher than that of stainless steel [49, 50]. However, uniqueness of materials still depends upon strength that makes them apart from others. Furthermore, carbon nanotubes are those materials that showed the strongest stiffness in the history of mankind. The tensile strength of carbon nanotubes measured so far is up to 63 GPa that is considered about 50 times greater than that of stainless steel [51]. However, carbon nanotubes that are identified as the weakest one show only several GPa strength [52]. As far as chemical, environmental stability, thermal conductivity etc. are compared to diamond. Owing to such attractive properties along with lightness of carbon nanotubes opens new routes towards variety of applications particularly in the field of aerospace [40, 53–55].

Carbon nanotubes highly exhibit electronic properties as compared to other materials. On comparing with copper carbon nanotubes show an extraordinary electrical conductivity. The most notable fact here is metallic as well as semiconducting nature of carbon nanotubes. The rolled-up structure comes forward to break up symmetric shape of the planar system. In this way, different directions are observed attributing to hexagonal lattice of carbon material and also axial direction is disturbed. Axial direction and unit vectors describe hexagonal lattice, therefore, depending on electrical properties carbon nanotubes may have nature of metal or semiconducting material. Amongst other nanotubes, semiconducting nanotubes may have band gap inversely with diameter. Band gap range was found between (1.8–0.18 eV) relative to small diameter tubes as well as very wide SWCNT respectively [56, 57]. Consequently, various nanotubes may belong to higher conductivity as compared to copper metal, while some others relative to silicon have a more conducting nature. There is a still promising interest in fabrication of nanoscale electronic devices by active use of nanotubes. Various areas of technology need carbon nanotubes to prepare advanced materials. Thus carbon nanotubes are already frequently used in those areas of research. Some outcomes of nanotubes are flat-panel displays, fuel cells, scanning probe microscopes, and sensing fabricated devices [58].

4.1 Optical properties

Electronic properties owing to SWNTs have been theoretically studied in early decades. SWNTs may be predicted metallic or semiconductors based on parameters that are followed in structure formation of the nanotubes [35]. As far as metallic and semiconducting nature of nanotubes is concerned, one third belongs to metallic whereas two-third relates to semiconducting nanotubes concerning selected indices (n, m). The aforesaid model is identified as π tight-binding model related to zone-folding scheme. Tight-binding data is based on (σ and π) bands that produce the curvature of σ and π bands. This bending behavior indicates a very small gap lying between metallic and semiconducting nanotubes [59, 60].

4.2 Electrical properties

Electrical properties of carbon nanotubes show electrical transport impact that becomes an interesting area of various possible applications attributing to fabricate electronic devices at nanoscale basis. Nanotubes are classified as one-dimensional conductor owing to which attractive microscopic phenomena are observed at low temperatures. Phenomena are likewise single-electron charging, superconductivity, and resonant tunneling. On the other hand, high temperature based tunneling conductance expresses power-law suppression that is evaluated as a function of (temperature and bias voltage) consistent with one-dimensional Luttinger liquid. Scattering mechanism is raised by optical or zone-boundary-phonons in metal-like nanotubes. Scattering along with coherent-backscattering phenomena has resulted in the form of low-temperature phenomena. Probe measurements were two-type as well as four-type in transport experiments performed with respect to MWNTs [61], isolated SWNTs, and SWNT bundles respectively [62, 63].

Initially, electrical resistance was measured towards unique MWNT below $T = 20$ mK, Langer et al. determined [61], whereas magnetic field shows a logarithmic conductance trend at declining temperature whereas saturation level was identified at the temperature below $T \sim 0.3$ K. However, when magnetic field impact was measured and found perpendicular towards tube axis, at that time magneto-resistance measurements were also observed. Furthermore, temperature effect on conductance in magnetic field was also observed that was found inconsistent with two-dimensional weak-localization.

4.3 Vibrational properties

Atomic-vibrations into carbon nanotubes were successfully evaluated by employing force-constant models (zone-folding-approximation) [64], also for concrete structure of nanotubes [65], ranging (tight-binding-models) [66–69] and finally, ab-initio models were also observed [70]. To measure vibrational eigenfrequencies, experiments were performed by using light resonant Raman scattering in case of laser-light-energy when energy measurements are very close to available electronic transitions. Resonance limitations are entirely different for all types of nanotubes; therefore Raman spectroscopy presents results to display various nanotubes structures that exist in the nanotube specimen. Currently, Raman spectroscopy measured parallel polarized light relevant to MWNTs [71], SWNTs [72, 73] and cross-polarized-light on isolated SWNTs [74].

4.4 Thermal properties

Phonons were used to measure specific heat as well as thermal conductivity of carbon nanotube systems. When temperature was kept low enough, acoustic phonons were observed indicating dominant role of phonon contribution in the nanotube systems. Linear specific heat measurements and thermal conductivity yield at or above 1 K but below room-temperature [75, 76], whereas 0.62 T specific heat identifies temperature at or below 1 K [77]. Linear temperature was evaluated depending on linear k-vector and modes of vibration of acoustic phonons such as longitudinal and twist like vibrations [78]. Transverse acoustic phonons are relative to specific heat exhibits dependence behavior attributing to specific heat at or below 1 K along with quadratic k-vector trend [79]. Thermoelectric measurement power (TEMP) for nanotube systems presents active and direct information about carrier types along with conductivity mechanisms [80–83].

5. Synthesis of CNTs

High-quality carbon nanotubes are considered to be superior quality materials and proved to be main pillar towards promising and versatile applications, various synthesis routes are employed to achieve feasible application of CNTs as described in **Figure 3**. Superior quality indicates that density of structural defects is significantly less over length scale between 1 to 10 microns along tube-axes. Carbon nanotubes synthesis is rapidly increasing in research field but still, challenges are prevailing. Those challenges are required to resolve with respect to synthesis of CNT. The main challenges are of four types regarding nanotube synthesis [84]. First is mass-production scale, containing low-cost based synthesis with large-scale synthetic routes to produce high-quality SWCNTs nanotubes. Second is a selective production scale that raises control over structural defects and changes electronic properties relevant to produced nanotubes. Third is Organization level regarding control over location along with specific orientation towards produced nanotubes on specific substrate. Fourth is mechanism level that presents all procedures followed during growth of nanotubes in synthetic processes. But growth mechanism is considered still controversial because alternative mechanisms may be employed during fabrication of CNTs [27, 85, 86].

Different techniques have been systematically employed to develop and produce SWNTs as well as MWNTs showing various structural and morphological characters in laboratory quantities. Methods commonly followed are three in number to synthesize CNTs, first is arc discharge [87, 88], second is laser ablation [66, 89] and third is chemical vapor deposition [67–69, 90, 91]. Catalysts are considered basic elements that are selected as source of carbon towards nanotubes formation, having sufficient energy. A significant feature of all methods followed for CNTs

Figure 3.
Currently used methods for CNTs synthesis [84].

fabrication is to enhance energy for carbon source producing fragments of carbon atoms that may recombine to yield SWNTs or MWNTs. The main goal is source of energy that is electricity and heat from an arc discharge and CVD respectively or high-intensity-light for laser ablation.

5.1 Arc discharge and laser vaporization

Amongst various methods that were allowed regarding SWNTs synthesis, arc-discharge or laser-ablation methods contributed relatively on large-scale basis (**Figure 4**). Subsequently, carbons atoms in a gaseous state are condensed caused by evaporation process of solid-state carbon atoms [92]. While growing single-wall-nanotubes (SWCNTs) in arc-discharge system, metallic catalyst is mandatorily required to incorporate for speed-up desired chemical reactions [93]. On the other hand, superior-quality (SWCNTs) are successfully fabricated (1–10 g scale) by using a laser oven approach [94]. Besides aforementioned method wave, CO_2-laser system was also employed regarding industrial-scale production of SWCNTs [95]. However, costly equipment as well as high energy consumption requirement makes them unfavorable approaches towards production of nanotube materials. Through employing arc ablation or laser methods only powder type specimens of carbon materials into bundle-shape form are controllably produced. The most common characteristic relevant to arc-discharge and laser-ablation approaches indicates higher energy need to induce carbon atoms to rearrange forming CNTs. Favorable temperature is prominently 3000 °C (or higher value) that is considered more beneficial for fine crystallization growth of CNTs at this level since products are obtained with attractive graphite-alignment. Moreover basic needs of the systems such as vacuum-conditions, repeated graphite-target substitution create barriers towards production of CNTs on an industrial scale [96].

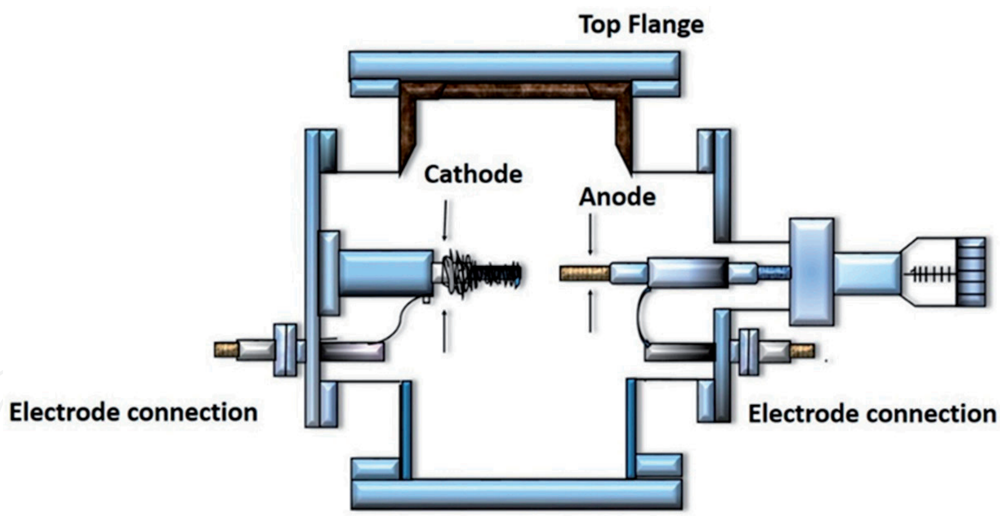

Figure 4.
Schematic diagram showing the Arc discharge method [40].

5.2 Chemical vapor deposition (CVD)

CVD approach presents carbon compounds decomposition in gaseous state where metallic nanoparticles are used as catalysts resulting in nucleation sites available for initial growth of carbon nanotubes (CNTs). Main drawback found in previous both methods was lack of large-scale fabrication of carbon materials, but CVD approach has presented preferred route towards carbon nanotubes production at large scale [97–99]. In this work, carbon is extracted from hydrocarbon source or some other carbon generating source. These chemical reactions are only successfully performed by using catalysts at or below 1200 °C temperature. Resultantly, CNT structure involved parameters like wall number, length, alignment, and diameter that have proven controllable CVD process. In addition, CVD approach has greater scope and advantages over other methods showing mild operation with low cost and selective process. The previous twelve years period describe that various approaches have presented promising industrial-scale synthesis of carbon nanotubes. All approaches indicate that CVD methods are main pillars of large scale production of nanotubes. Among various methods, main approaches are five in numbers that have proven to be successful large-scale yield [100].

i. Methane(CH_4) chemical vapor deposition (CVD) approach has been reported in 1998 showing bulky synthesis of SWCNTs by employing CVD method directly from CH_4 at temperature level (900 °C) [101, 102]. But Su *et al*. [103] successfully enhanced yield efficiency of CVD method by using aluminum oxide (Al_2O_3) aerogels. These aerogels were impregnated with a catalyst containing Fe/Mo nanoparticles.

ii. High-Pressure carbon monooxide (HPCO) approach presented catalytic-decomposition of CO through CO (a carbon source) at high pressure towards SWCNTs fabrication [104]. Aforesaid catalysts were used in decomposition process in the form of a gas phase state collected from organometallic-catalyst that was used as a precursor.

iii. In CO-CVD approach CO gas plays role to feed gas. On comparing with previously prepared samples with same methane catalyst, amorphous carbon rate was drastically reduced. In addition, the use of Co-Mo catalyst was considered an additional advancement in previously employed CO-CVD approach [105]. This approach incorporates Co/Mo bimetallic catalyst along with fluidized CVD-reactor during production of SWCNTs at large scale. The main benefit that arises from controllable use of fluidized CVD reactors was that they stop continuous addition as well as removal of solid-like particles during operation without stopping reactor work.

iv. Alcoholic CVD approach has been presented by Maruyama *et al*. in 2002 report [106] by producing superior quality SWCNTs with aless amount of amorphous carbon. Alcohols were used in methanol or ethanol from carbon source. Resultantly, formation of OH radical during synthesis process played a vital role for maximum removal of amorphous carbon from efficient growth of pure SWCNTs leaving them advanced material in a research field.

v. Currently Plasma Enhanced Chemical Vapor Deposition (PECVD) approach has been used widely towards fabrication of MWCNTs and SWCNTs carboneous compounds [107–109]. Moreover reactive species present in plasma-system may seriously affect growth of carbon nanotubes with small diameter, thereby creating implications for diameter control along with selective etching attributing to metallic SWCNTs growth.

5.3 Laser ablation method

Both laser ablation and arc discharge approaches have the same principles with similar mechanisms. However, they are not similar with respect to energy sources that are adopted to complete reactions. A laser is main source of desired energy for laser ablation method and **Figure 5** is showing schematic experimental setup. The schematic structure contains quartz tube with graphitic block. Graphite block is heated at 1200°C temperature by using high-power-laser whereas metal particles are incorporated catalysts [110]. Argon gas is controllably used in stream form during reaction process. Graphite lying in quartz is systematically vaporized by functioning of laser. Argon present in chamber removes vapors of carbon by condensation process towards downstream cooler quartz-walls. Condensation process is completed in the presence of both SWCNTs and metallic nanoparticles (see **Figure 5**). Literature reveals that laser power may

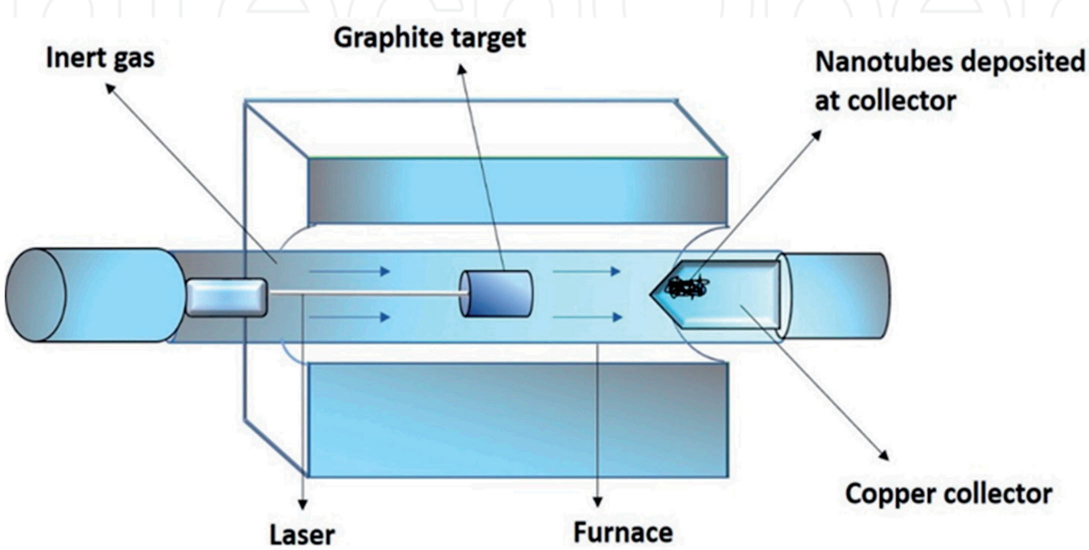

Figure 5.
Schematic structure showing the laser ablation method.

strongly affect CNTs diameter. On increasing laser pulse power rate a very thin diameter carbon nanotube is collected [40]. On other hand, some other reports give more information in favor of laser pulses that they behave like great potential, owing to be capable to provide SWCNTs in large amount [89, 111]. Reports indicate pure and superior quality production of SWCNTs in this case. Curved graphene sheets are observed showing position of carbon atoms in condense phase state caused by set up created by metal-catalyst to fabricate condensed carbon nanotubes.

In this case, carbon atoms rearrange them for formation of ring shape and in this way, electronegative properties become dominant to play role in preventing open edge from sealing [110]. Furthermore, there are main benefits relative to this method that indicate metallic impurities less in amount but high in yield owing to vaporization tendency creating at tube end of metallic atoms at closing position. However main drawback of technique is observed with respect to synthesis aspect of nanotubes that they are not regularly straight rather indicate degree of branching to some extent. In addition this technique involves high-quality graphitic rods with high-power laser rate. However, in this case, CNTs are produced but not greater than arc-discharge technique. Carbon-2019 for "PEER REVIEW" describes high-power-laser when metal particles are incorporated as mandatory catalysts in reaction process [110]. Argon gaseous stream is continuously used during reaction mechanism. Graphitic quartz is passed through vaporization process using a laser, argon media captures carbon vapors that result in condensed downstream towards cooler-walls of quartz but still SWCNTs with metallic-particles are located in condensation process. Laser power may also clearly affect CNTs diameter. Furthermore, diameter becomes comparatively narrowed on increasing laser pulse rate [30]. Other studies reported that ultrafast laser pulses are of great potential, and are capable to produce larger quantities of SWCNTs [112]. SWCNTs collected by this technique are observed owing to high-purity and superior-quality in nature. Location sites where carbons atoms initiate condensation process may set up curved shape graphene sheet along with metal-catalyst atoms. In this way condensed nanotubes are properly obtained showing peculiar properties. Moreover, carbon atoms merge to form specific rings, thereby raising electronegative properties relative to metallic atoms that become capable to prevent open-edge from closing [113].

The main benefit belongs to followed method, in this case, metallic impurities are observed relatively less in amount but with high yield that is caused by vapors formation tendency belonging to metallic atoms from tube end when closed once in a time. However main drawback relative to this technique indicates irregularity in straight shape for synthesized nanotubes whereas degree of branching occurs to some extent. Furthermore, pure graphitic rods are involved in this procedure along with high laser power rate. Resultantly production of CNTs was not in great amount as compared to arc- discharge method.

6. Conclusions

Carbon nanotubes have the ability to be more investigated, and it is possible to drive further advancements by using CNTs in different fields. The findings obtained in the synthesis, functionalization, and structure of CNTs have contributed significantly to promising developments in various fields. However, further perfections in synthesis protocols are needed to obtain highly durable CNTs for preferred applications. For an instant, catalyst size is directly influenced on diameter of CNT during CVD reaction. So, further analysis should also be undertaken to discover more effective methods of processing precisely uniform-sized catalyst particles in order to ensure the production desired diameter of SWCNTs; but CNTs are costly than other carbon nanomaterials. Efforts should be proceeded to look for modern, cost-effective, and plentiful carbon sources, so that cost of CNTs can be lowered to an acceptable amount.

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