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CARBON NANOTUBES AND THEIR APPLICATION IN BIO-SYSTEMS

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ABSTRACT:

Carbon nanotubes (CNTs) are allotropes of carbon with a nanostructure that can have a length-to-diameter ratio greater than 1,000,000. These cylindrical carbon molecules have novel properties that make them potentially useful in many applications in nanotechnology. Nanotubes are categorized as single-walled nanotubes, multiple walled nanotubes, torus, nanobuds, cup shaped carbon naotubes and functionalized CNTs. Techniques have been developed to produce nanotubes in sizeable quantities, including arc discharge, laser ablation, chemical vapor deposition, silane solution method and natural, incidental, application related issues, and controlled flame synthesis. The properties of CNTs are still being researched heavily and scientists have barely begun to tap the potential of these structures. They can pass through membranes, carrying therapeutic drugs, vaccines and nucleic acids deep into the cell to targets previously unreachable.

Key words: Carbon nanotubes, torus, f-CNTs, arc discharge, laser ablation, chemical vapour deposition.

INTRODUCTION:

Nanostructures (tubes, rods, crystals, sheets, etc.) are one of the most exciting topics in modern materials chemistry, and have a dizzying array of applications in areas as diverse as clinical medicine, industrial catalysis, and consumer electronics. By far the most popular and well known of all nanostructures is the carbon nanotube[1], such novel nanomaterials consist of inorganic or organic matter and in most cases have never been studied in the context of pharmaceuticals, carbon nanotubes (CNTs) are one of them. They are nanometers in diameter and several millimeters in length and have a very broad range of electronic, thermal, and structural properties. These properties vary with kind of nanotubes defined by its diameter, length, chirality or twist and wall nature. Their unique surface area, stiffness, strength and resilience have led to much excitement in the field of pharmacy. CNTs are allotropes of carbon. They are tubular in shape, made of graphite. CNTs possess various novel properties that make them useful in the field of nanotechnology and pharmaceuticals[2]. Carbon nanotubes could be visualized as rolled sheets of graphene (sp2 carbon) that are sometimes capped at each end. They could be either single-walled with diameters as small as about 0.4 nm, or multi-walled consisting of nested tubes (2–30 concentric tubes positioned one inside the other) with outer diameters ranging from 5 to 100 nm. The electronic properties of single-walled carbon nanotubes (SWNTs) may vary from semiconductors to metals, depending upon the chiral angle (the way the hexagons are positioned with respect to the tube axis[3].

HISTORY:

In 1952 Radushkevich and Lukyanovich published clear images of 50 nanometer diameter tubes made of carbon in the Soviet Journal of Physical Chemistry. A paper by Oberlin, Endo, and Koyama published in 1976 clearly showed hollow carbon fibres with nanometer-scale diameters using a vapor-growth technique. Furthermore, in 1979, John Abrahamson presented evidence of carbon nanotubes at the 14th Biennial Conference of Carbon at Penn State University. The conference paper described carbon nanotubes as carbon fibers which were produced on carbon anodes during arc discharge. In 1981 a group of Soviet scientists published the results of chemical and structural characterization of carbon nanoparticles produced by a thermocatalytical disproportionation of carbon monoxide. Using transmission electron microscopy (TEM) images and X-ray diffraction (XRD) patterns, the authors suggested that their “Carbon multi-layer tubular crystals” were formed by rolling graphene layers into cylinders. In 1987, Howard G. Tennent of Hyperion Catalysis was issued a U.S. patent for the production of "cylindrical discrete carbon fibrils" with a "constant diameter between about 3.5 and about 70 nanometers, length 10^2 times the diameter, and an outer region of multiple essentially continuous layers of ordered carbon atoms and a distinct inner core.

A large percentage of academic and popular literature attributes the discovery of hollow, nanometer sized tubes composed of graphitic carbon to Sumio Iijima of Nippon Electric Company in 1991. A 2006 editorial written by Marc Monthioux and Vladimir Kuznetsov in the journal Carbon has described origin of the carbon nanotube [4, 5].

STRUCTURE AND MORPHOLOGY

Carbon nanotubes are allotropes composed entirely of carbon in the form of a hallow sphere, ellipsoid, or tube. CNTs are sp^2 bonded, with each atom joined to three neighbours, as in graphite. Thus, tubes are considered as rolled-up graphene sheets [6].

CLASSIFICATION OF CARBON NANOTUBES

1. Single-walled
2. Multi-walled
3. Double-walled
4. Torus
5. Fullerene
6. Nanobud
7. Functionalized CNTs

Single-walled Nanotubes:

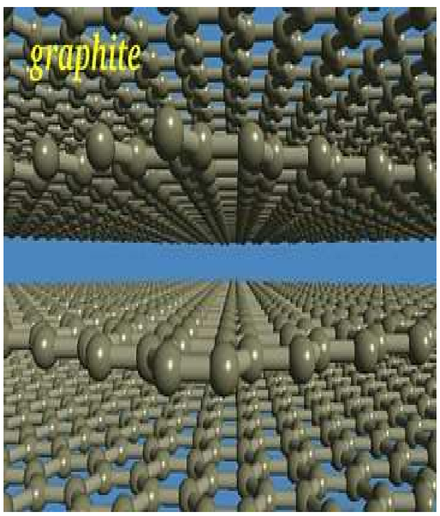
Single walled nanotubes (SWNTs) are proposed to consist of single cylindrical graphitic sheet capped by hemispherical ends composed of pentagons and hexagons. Curves observed in high-resolution transmission electron microscope (HRTEM) images of SWNTs[7].

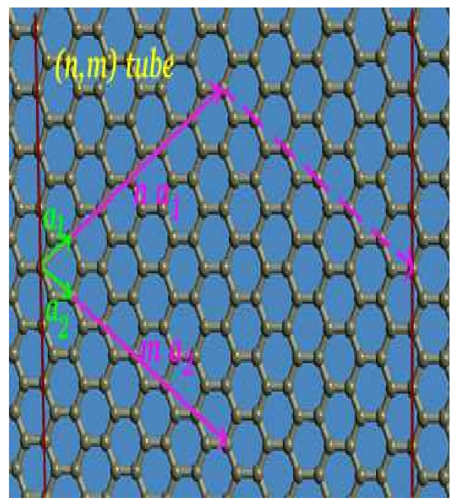
Graphite consists of layers of carbon atoms. Within the layers the atoms are arranged at the corners of hexagons which fill the whole plane (in the idealized case without defects). The carbon atoms are strongly (covalently) bound to each other (carbon-carbon distance ~ 0.14 nm), leading to a very large in plane value for Young's modulus.

The layers themselves are rather weakly bound to each other (weak long range Vander Wall type interactions, interlayer distance of ~0.34 nm).

The weak interlayer coupling gives a very soft nature to graphite.

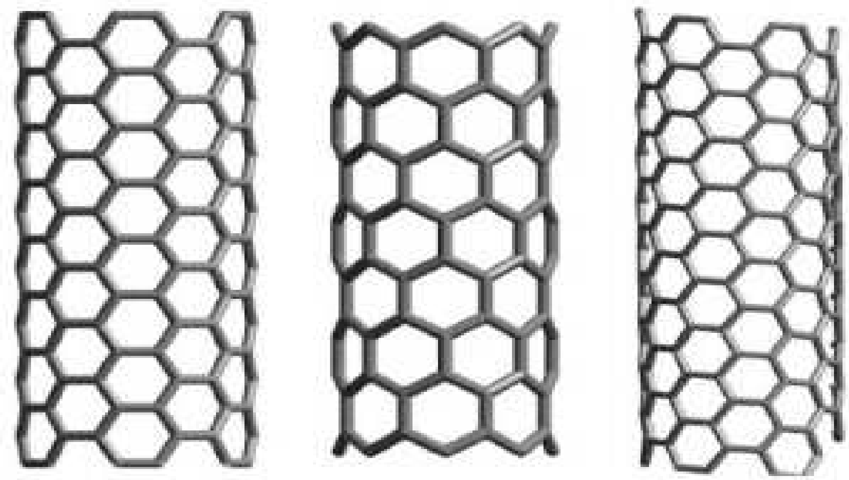
To build a nanotube, we in mind, take out, one single layer from the graphite stack and wrap it to cylindrical shape in the following way: from the plane we cut out a slice (marked by the left and right dark red vertical lines in the figure) in a way, so, if wrapped into a cylinder, atoms on the left dark red line can be mapped onto atoms on the right dark red line. This mapping condition ensures that, again, only hexagons can be found on the surface of the cylinder. Starting with only one layer of 2-dimensional graphite we end up with a cylinder with only one wall, a single wall carbon nanotube[8].(figure:1,2)





1. Single layer of graphite stack **2. Graphite showing layers of "C"atom warp into CNT**

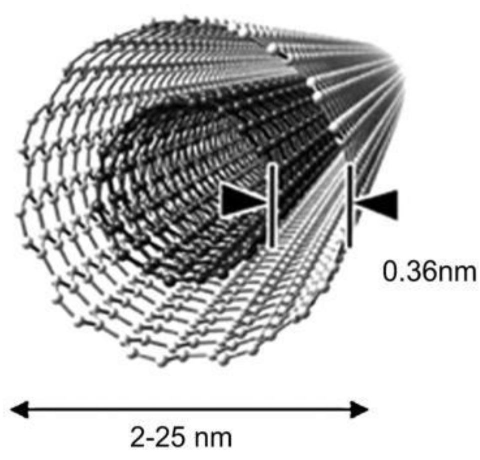
Single walled nanotubes (SWNT) roughly have a 1 nanometer diameter, with a tube length that can be up to 28 million times longer. The structure of a SWNT can be conceptualized by wrapping a one-atom-thick layer of graphite called graphene into a seamless cylinder. Depending on their structure the SWNT are named *zigzag*, *armchair*, *graphene nanoribbon* or *chiral* [9].(figure:3)



3. Single walled CNTs structures

Double-wall Nanotubes (DWNT)

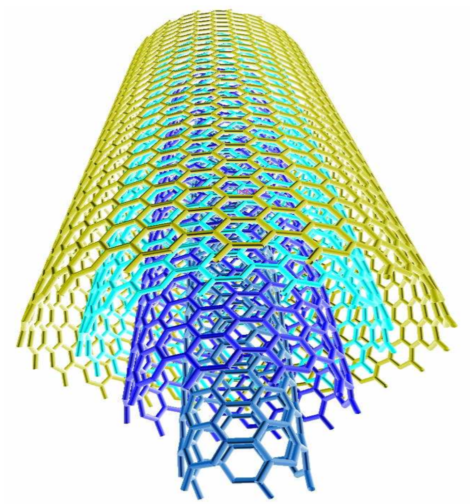
These materials combine similar morphology and other properties of SWNT, while significantly improving their resistance to chemicals. Double-wall nanotubes are ideal systems for studying the interwall interactions influencing the properties of nanotubes with two or more walls. This property is especially important when functionality is required to add new properties to the nanotube. Since DWNT are a synthetic blend of both SWNT and MWNT, they exhibit the electrical and thermal stability of the latter and the flexibility of the former [10, 11].(figure:4)



4**. Double walled carbonnano tube**

Multi-wall Nanotubes (MWNT)

Multi-wall carbon nanotubes (MWCNT) obtained by catalytic decomposition of iron phthalocyanine are investigated by high resolution electron microscopy and electron diffraction (ED)[12]. Multi-wall nanotubes can appear either in the form of a coaxial assembly of SWNT similar to a coaxial cable, or as a single sheet of graphite rolled into the shape of a scroll. The diameters of MWNT are typically in the range of 5 nm to 50 nm. The interlayer distance in MWNT is close to the distance between graphene layers in graphite[13].(figure:5)



5. Multi walled carbonnano tube

Torous:

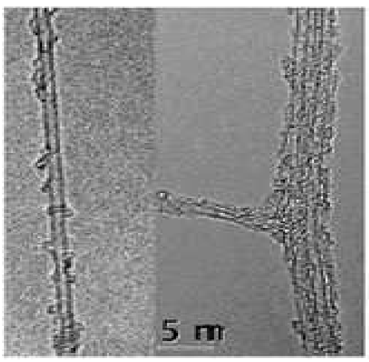
Carbon nanostructures have been synthesized in NaCl-$MgCl_2$ and in NaCl-$CaCl_2$ salt melts and the extracted material was investigated by tapping-mode atomic force microscopy (TM-AFM) and scanning electron microscopy. Some interesting new nanostructures were found and investigated as torus-shaped carbon structures with a ring diameter of 300-400 nm and 10-15 nm height. These tori are closely related to the wrapped SWNT rings described recently. They are probably formed during the electrolysis[14].

Fullerene:

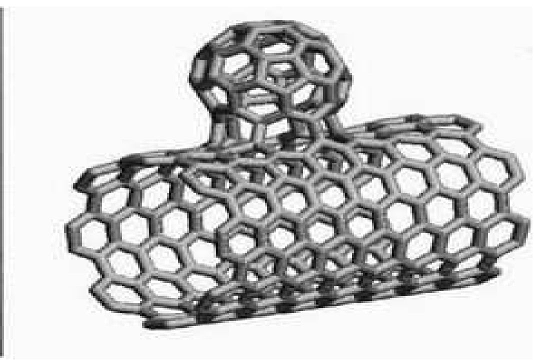
A fullerene is molecule composed entirely of carbon, in the form of a hollow sphere, ellipsoid, or tube. Spherical fullerenes are also called buckyballs, and cylindrical ones are called carbon nanotubes or buckytubes. Fullerenes are similar in structure to graphite. The first fullerene to be discovered, and the family's namesake, was buckminsterfullerene C_{60}, made in 1985 by Robert Curl, Harold Kroto and Richard Smalley. The name was homage to Richard Buckminster Fuller, whose geodesic domes it resembles. Fullerenes have since been found to occur in nature[15].

Nanobud:

A new material, the carbon nanobud, has been developed at Helsinki University of technology, in Finland[16]. The newly discovered structures, dubbed Nanobuds because they resemble buds sprouting on branches, are a hybrid carbon nanomaterial that merges the single-walled nanotubes, SWNT, and fullerenes it may possess properties that are superior to fullerene and nanotube alone.(figure:6,7).



6. TEM image of Nanobud



7. Fullerene attached to SWCNT

The creation of the Nanobuds began with the synthesis of single-walled carbon nanotubes in a standard reactor. The resulting SWNTs seemed to be coated with clusters of carbon atoms, but a closer investigation, using an electron microscope, revealed that most of the clusters were actually fullerenes.

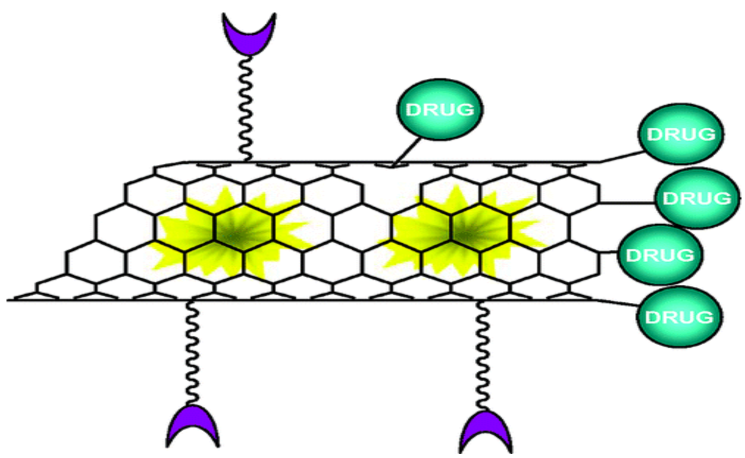
This deviation from the characteristic fullerene sphere, or "bucky ball," could have been due to the presence of covalently attached oxygen or hydrogen. Further measurements detected oxygen in each Nanobud, and a round of infrared spectroscopy revealed the presence of two types of organic compounds known as ethers and esters. These compounds may act as bridge-like structures connecting the fullerenes to the nanotubes.

Nanobuds could have many applications as they may find use as memory devices and quantum dots or cold electron field emitters-materials that emit electrons at room temperature under a high applied electric field-due to the fullerenes many curved surfaces, which make for better emitters than flat surfaces. Cold electron field emission is the key to many technologies, including flat-panel displays and electron microscopes[17, 18].

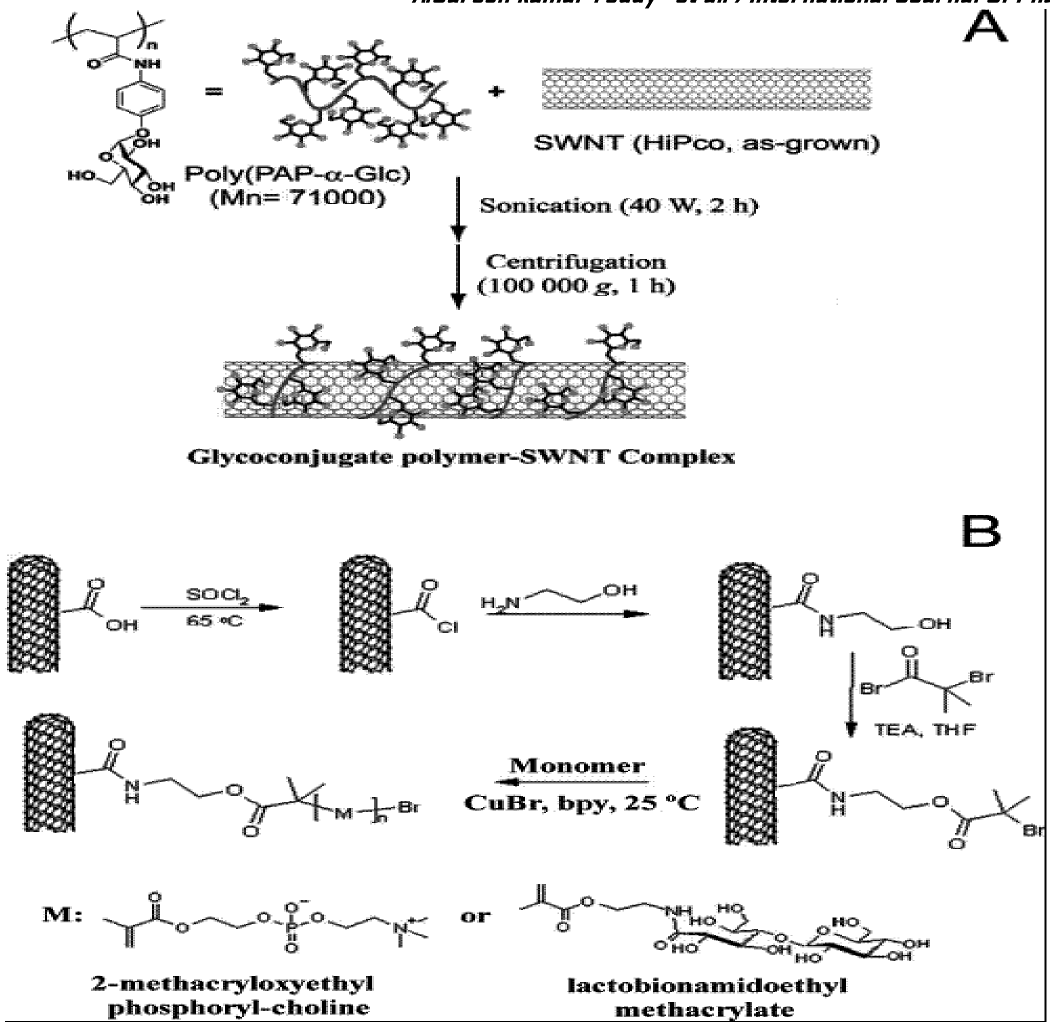
Functionalised carbon nanotubes:

Functionalised carbon nanotubes are which contain additional functional groups on their surface. They have superior properties that normal carbon nanotubes. Carbon nanotubes when treated with mixtures of concentrated sulfuric and nitric acids, they result in the formation of carboxyl and hydroxyl groups on their surface. These activated CNTs are able to react with other functional groups favoring coupling to different compounds[19].

Single walled carbon nanotubes (SWCNTs) are functionalized using molten urea as the solvent, and dispersed with arenediazonium salts in less than 15 min [20].(figure:8,9)



8. Funtionalised CNTs loaded with drug



9.Funtionalised CNTs

A. SWCNTs wrapped by glycol-conjugate polymer with bioactive sugars.

B. Modification of carboxyl-functionalized SWCNTs with biocompatible, water-soluble phosphorylcholine sugar-based polymers.

SYNTHESIS:

1. Arc discharge
2. Laser ablation
3. Chemical vapor deposition (CVD)
4. High pressure carbon monoxide (HiPCO)
5. Others

i) Natural

ii) Incidental

iii) Controlled flame environments

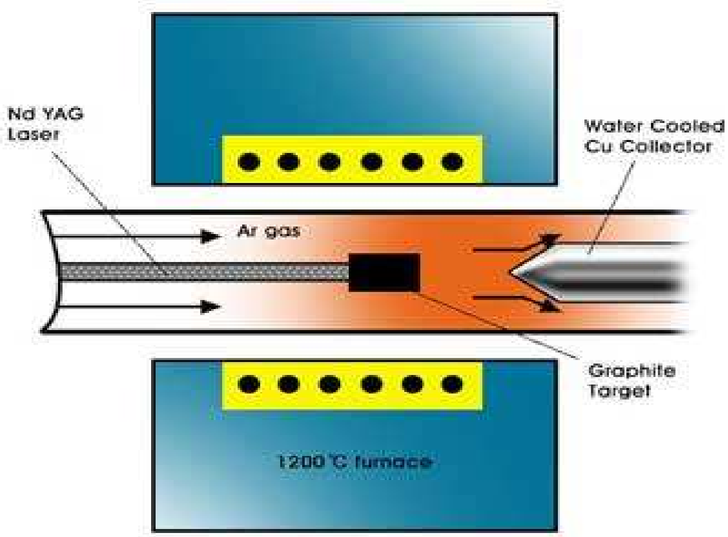
Arc discharge method:

The carbon arc discharge method initially used for producing C_{60} (fullerenes), is the most common and perhaps easiest way to produce CNTs, as it is rather simple. However, it is a technique that produces a complex mixture of components, and requires further purification to separate the CNTs from the soot and the residual catalytic metals present in the crude product. This method creates CNTs through arc-vaporization of two carbon rods place end to end, separated by approximately 1mm, in an enclosure that is usually filled with gas such as he, H_2, N_2, O_2, C_2H_2, and CO_2 and at different pressures (3-100Kpa). Recent investigations have shown that it is also possible to create CNTs with the arc method in liquid nitrogen. A direct current of 50 to 100A, driven by a potential difference of approximately 20V, creates a high temperature discharge between the two electrodes. They vaporize the surface of one of the carbon electrodes, and forms a small rod-shaped deposit on the yield depends on the other electrode. Producing CNTs in high yield depends on the uniformity of the plasma arc and the temperature of the deposit forming on the carbon electrode [21, 22, 23].

Laser ablation:

In 1995 Smalley's group at Rice University reported the synthesis of carbon nanotubes by laser vaporization. The laser vaporization apparatus used by Smalley's

group is shown in figure. A pulsed or continuous laser is used to vaporize a graphite target in an oven at 1200°C. The main difference between continuous and pulsed laser, is that the pulsed laser demands a much higher light intensity (100 KW/cm^2 compared with 12 KW/cm^2)[24]. Samples were prepared by laser vaporization of graphite rods with a 50:50 catalyst mixture of cobalt and Nickel at 1200°C in flowing argon, followed by heat treatment in a vacuum at 1000°C to remove the C_{60} and other fullerenes. The initial laser vaporization pulse was followed by a second pulse, to vaporize the target more uniformly. The use of two successive laser pulses minimizes the amount of carbon deposited as soot. The second laser pulse breaks up the larger particles ablated by the first one, and feeds them into the growing nanotube structure. The material produced by this method appears as a mat of "ropes", 10-20 nm in diameter and up to 100 µm or more in length. Each rope is found to consist primarily of a bundle of single walled nanotubes, aligned along a common axis. By varying the growth temperature, the catalyst composition, and other process parameters, the average nanotube diameter and size distribution can be varied[25]. CNTs are preferentially collected by electro-statically biasing (5 kVDC) the substrates[26].(figure:10).



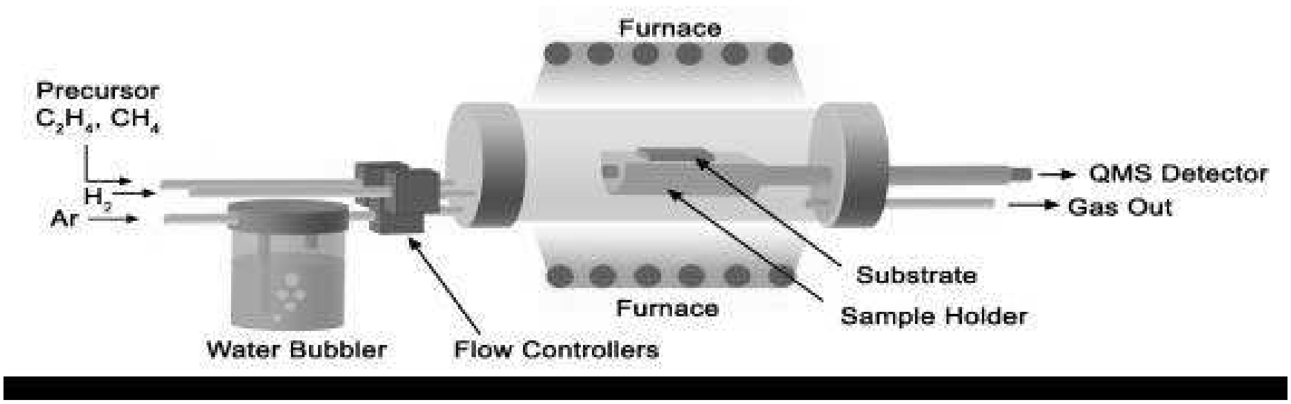
10. Synthesis of carbo nano tubes by laser ablation method.

Chemical vapour deposition (CVD):

Chemical vapour deposition CVD is known as irreversible deposition of a solid from a gas or a mixture of gases through a heterogeneous chemical reaction. This reaction takes place at the interface of gas-solid substrate, and depending on the deposition conditions, the growth prosess can be controlled either by diffusion or by surface kinecitcs. This process easily scaled up to industrial production. CVD is a continuous process and currently is the best- known techniuque for high yield, and low impurity production of CNT at moderate temperatures. The CVD technique has become one of the preferred methods for fabricating carbon nanotubes in much lower temperature regimes than is possible with the arc-discharge technique. CVD methods utilize the pyrolytic decomposition of hydrocarbon gases at elevated temperatures in the range 600–1200°C. Several researchers have reported successful syntheses of carbon nanotubes using CVD techniques recently. For example, it has been reported that the observation of nanotubes in the pyrolytic product of benzene (C_6H_6) decomposition at about 1100°C. The synthesis of carbon nanotubes was report at about 750°C using a CVD technique.

$CH_4(g)$. $H_2(g)$ → 600----1200 `C Ferrocene C(s) . $3H_2(g)$

At high temperatures the bonds in methane are broken the free carbon atoms attached to the catalyst where it bond to other carbon atom. Catalyst Fe, Co, and Ni are frequently used other metals such as Sc, Ti, V, Cr, Mn, Y, Zr, Nb, Mo, Hf, Ta, W, Re and combination of them [27, 28].(figure:11).



11. Synthesis of carbon nano tube by chemical vapour deposition method.

High Pressure CO disproportionation (HiPCO):

The High Pressure CO disproportionation process (HiPCO) is a technique for the catalytic production of SWNTs in a continuous-flow gas phase, using CO as the carbon feedstock and Fe $(CO)_5$ as the iron-containing catalyst precursor. SWNTs are produced by flowing CO, mixed with a small amount of Fe $(CO)_5$, through a heated reactor. Figure 3 shows the layout of the CO flow-tube reactor. Size and diameter distribution of the nanotubes can be roughly selected by controlling the pressure of the CO. This process is promising for bulk production of carbon nanotubes. The average diameter of HiPCO SWNTs is approximately 1.1 nm. The highest yields and narrowest tubes can be produced at the highest accessible temperature and pressure [29].

APPLICATIONS OF CARBON NANOTUBES IN BIO – SYSTEMS

- Diagnosis
- Drug discovery
  - Delivery

- Peptides
- Proteins
- Nucleic acids
- drugs

➢ Tissue Engineering

Diagnosis:

1. Carbon nanotubes can also be employed as a powerful carrier to pre-concentrate enzymes or electroactive molecules for electrochemical sensing of DNA hybridization as a novel indicator [30].
2. Multiwalled carbon nanotubes functionalized with europium-doped Y_2O_3 nanophosphors gives rise to species that are luminescent in the visible-light range analysed by *Z*-contrast imaging and such species have potential applications in cancer diagnosis and treatment[31].
3. Carbon nanotubes improve cancer diagnosis through better protein array detection limits[32].
4. In recent trends biological Functionalized carbon nanotubes have their potential applications for breast cancer diagnostic[3].
5. Carbon nanotube-filled, nanocomposite-derived catheter exhibited outstanding properties when compared with neat polymer-derived catheters, and it is envisaged that these system will be widely utilized in various medical devices.
6. Nanotube-filled microcatheters were confirmed by measuring the systematic T-cells as well as a histopathological[33].

Drug discovery:

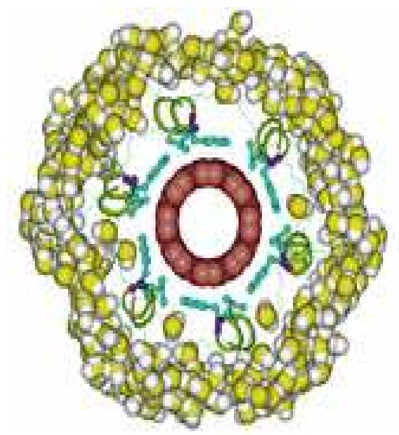
1. The critical bottlenecks in drug discovery may be overcome by using arrays of CNT sensors and current information technology solutions for identification of genes and genetic materials for drug discovery and development[34].

2. The versatile physicochemical features enable the covalent and noncovalent introduction of several pharmaceutically relevant entities and allow for rational design of novel candidate nanoscale constructs for drug development[35].

Delivery:

Peptide:

1. CNTs conjugated with antigenic peptides can be developed as a new and effective system for synthetic peptide applications[36].
2. Functionalized single wall carbon nanotubes (f-SWCNT) complexed with nanochitosan (NG042) and used for delivery of DNA encoding FITC-labeled peptide. F-SWCNT-chitosan is more effective in intracellular delivery of peptide compared to chitosan[37].(figure:12).



12. Delivery of peptides through CNTs

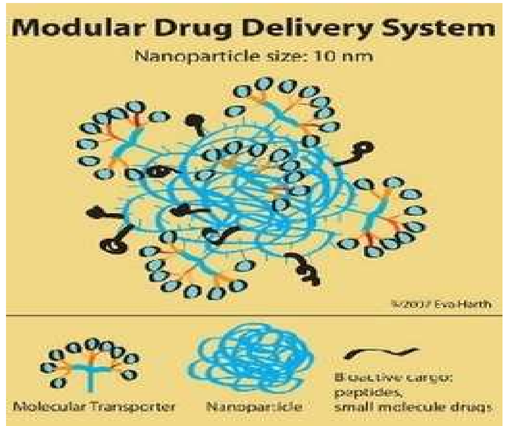
Proteins:

1. Researchers describe antigen-antibody interactions and immune responses using protein-carbon nanotube complexes[38].
2. Single walled carbon nanotubes with various functionalizations are capable of the transportation of proteins and oligonucleotides into living cells[39].

Drug:

1. Carbon nanotubes (CNT) have emerged as a new alternative and efficient tool for transporting and translocating therapeutic molecules. CNT display low toxicity and are not immunogenic[40].

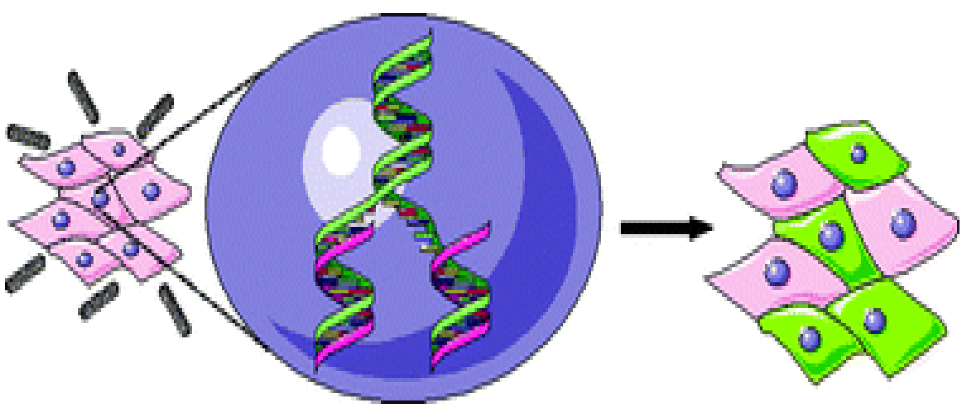
2. Combination of polymer technology and carbon nanotubes array act as effective drug delivery system at cellular levels[41].(figure:13).



13. Delivery of Drugs through CNTs

Nucleic acid:

4. Recent findings of improved cell membrane permeability for carbon nanotubes would expand medical applications to therapeutics using carbon nanotubes as carriers in gene delivery systems[42].
5. Functionalized, positively charged, water-soluble carbon nanotubes are able to penetrate into cells and can transport plasmid DNA by formation of noncovalent DNA–nanotube complexes. Such nanotubes can be used as novel nonviral delivery systems for gene transfer[43].(figure:14).



14. Nuclicacid delivery by carbon nanotubes.

6. Ammonium-functionalized CNTs can also be considered very promising vectors for gene-encoding nucleic acids. Indeed, we have formed stable complexes between cationic CNTs and plasmid DNA and demonstrated the enhancement of the gene therapeutic capacity in comparison to DNA alone[35].

Tissue engineering:

1. Polyurethane foams with CNT coating have the potential to be used as bioactive scaffolds in bone tissue engineering due to their high interconnected porosity, bioactivity and nanostructured surface topography[44].
2. Carbon nanotubes can also be incorporated into scaffolds providing structural reinforcement as well as imparting novel properties such as electrical conductivity into the scaffolds may aid in directing cell growth[45].
3. In cardiovascular surgeries remarkable improvements in mechanical strength of implanted catheters is brought by carbon nanotubes and reduce the thrombogenecity after surgery[46].
4. Though challenges still exist, the addition of CNT to improve the mechanical properties of CTS and ceramic (HAp) composite would surely support and stimulate the function of natural bone[47].

Case study:

1. Carbon nanotubes are used in trans-dermal drug deliver of nicotine[48].
2. Boron nitride nanotube provides the most ideal delivery capsule in terms of minimizing the amount of material required for encapsulation, this provides the least toxicity the technique used in encapsulation of cisplatin entering carbon[49].
3. Presence of different functional groups on the single-walled carbon nanotubes influences the anti-oxidant activity or free radical scavenging activity[50].
4. Water soluble conjugate SWCNT-PTX is formed by ester bond between Single Walled Carbon Nanotubes and Paclitaxel (PTX) is used as chemotherapy for tumor-targeting with high efficacy[51].
5. Researchers from the Universsity of Southern California have shown that CNTs may be used to model the functioning of brain cells (neurons). So this may lead to the creation of better artificial neurons and

potentially enable the construction of a synthetic electronic brain. Scientists have only tested computer simulations to see whether it is theoretically possible are not[52].

6. Researchers at Stanford University have used microelectrode arrays consisting of multi-walled carbon nanotubes to electrically stimulate rat retinal neurons, and developing bio-microelectromechanical systems (bioMEMS) devices for the treatment of age-related macular degeneration and glaucoma[53].
7. Anodized titanium covered by carbon nanotubes could lead to a new material for orthopedic implants[54].

CONCLUSION:

The science of nanomedicine has become an ever-growing field that has an incredible ability to bypass barriers. The properties of CNTs are still being researched. Functionalized carbon nanotubes have already proven to serve as safer and more effective alternatives to previous drug delivery methods and as diagnostic tool. They can pass through membranes, carrying therapeutic drugs, vaccines, and nucleic acids deep into the cell to targets previously unreachable. They also serve as ideal non-toxic vehicles which, in some cases, increase the solubility of the drug attached, resulting in greater efficacy and safety. Overall, recent studies regarding CNTs have shown that they can be used in artificial neurons.

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