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Review Article

Insight into Nanotechnology and Applications of Nanomaterials

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ABSTRACT

The evolution of technology and instrumentation as well as its related scientific areas such as physics and chemistry are making the research on nanotechnology aggressive and evolutionary. The ability to construct tiny objects atom by atom or molecule by molecule forms one of the exciting prospects of the research field in nanoscience. It shows great promise for providing us in the near future with many breakthroughs that will change the direction of nanotechnology advances in a wide range of applications. At the nanoscale dimensions, the material properties change significantly differing completely from their bulk counterparts. Nanomaterials display new phenomenon associated with the quantized effects and with the preponderance of surfaces and interfaces. Metal nanoparticle dispersions exhibit colors due to the surface plasmon resonance phenomenon, which is caused by the coherent oscillation of conduction band electrons when they interact with electromagnetic field. Carbon nano tubes contain two and more coaxial cylinders, respectively, with each cylinder being a single walled carbon nanotube with weak van der Waal forces binding the cylinders together. Many synthetic methodologies for zero and one dimensional nanostructures have been developed, including vapor phase methods and aqueous methods. Laser ablation has been extensively used for the preparation of nanoparticles and particulate films. The main aim of nanotechnology is to enhance productivity through nanotech driven precision farming and to maximize the output and minimize inputs through better monitoring and targeted.

Keywords: Nanoscience, Nanomaterials, Classification, Properties, Synthesis.

1. INTRODUCTION

The term "Nanotechnology" was first defined by Norio Taniguchi of the Tokyo Science University in 1974. Nanotechnology, shortened to "Nanotech", is the study of manipulating matter on an atomic and molecular scale. Generally nanotechnology deals with structures sized between 1 to 100 nm and involve developing materials or devices within that size. For comparison, 10 nanometers is 1000 times smaller than the diameter of a human hair. Nanotechnology has the capacity to improve our ability to prevent, detect and remove environmental contaminants in air, water and soil in a cost effective and environmentally friendly manner. Nanoscience and nanotechnologies are revolutionizing our understanding of matter and are likely to have profound implications for all sectors of the economy, including agriculture and food, energy production and efficiency, the automotive industry, cosmetics, medical appliances and drugs, household appliances, computers and weapons^{F-3}. This capability also simultaneously gives us the ability to build materials

and devices or shapes and products on that scale. Because of the brevity in operation, smarter and lighter products can be made from the molecules of the same matter with every atom in its specified place through 'positioned assembly' or 'self assembly'. The matter displays unimaginably different qualities when manipulated and structured at nanoscale. It produces different products when assembled at that scale. This is what is the future unleashed by the nanotechnology revolution. We have been still dealing with the matter at a bigger scale. The building blocks, our engineering skills and products were bigger than the nanosize and hence had limitations in manipulation. It is this arrangement of atoms which defines the properties of matter. With its capacity to manipulate the smallest possible component of the matter, the nanotechnology has the potential to bring that cycle of technological revolution to completion: dealing with the matter atom by atom, molecule by molecule. It is this capacity of mankind to deal with matter at molecular level that will give the

mankind a historical new ability to shape, process and create things which have never been thought of.

Nanotechnology, introduced almost half century ago, is one of the most active research areas with both novel science and useful applications that has gradually established it in the past two decades. The evolution of technology and instrumentation as well as its related scientific areas such as physics and chemistry are making the research on nanotechnology aggressive and evolutionary. Not surprisingly, it is observed that expenditure on nanotechnology research is significant. The U.S. National Nanotechnology Initiative (NNI) expenditures exceed \$1 billion each year, with the President's 2008 budget for NNI at \$1.5 billion. However, the research is mainly moving forward motivated by immediate profitable return generated by high value commercial products⁴. The branch of nanoscience and technology is truly multi-disciplinary and is an emerging technology with full of promises to have an impact on virtually every spectrum of civilization including communications, computing, textiles, cosmetics, sports, therapy, automobiles, environmental monitoring, fuel cells and energy devices, water purification, food and beverage industry, etc. The ability to construct tiny objects atom by atom or molecule by molecule forms one of the exciting prospects of the research field in nanoscience. It shows great promise for providing us in the near future with many breakthroughs that will change the direction of nanotechnology advances in a wider range of applications⁵⁻⁷.

Metal oxides play a very important role in many areas of chemistry, physics and materials science. The metal elements are able to form a large diversity of oxide compounds. These can adopt a vast number of structural geometries with an electronic structure that can exhibit metallic, semiconductor or insulator character. In technological applications, oxides are used in the fabrication of microelectronic circuits, sensors, piezoelectric devices and fuel cells, coatings for the passivation of surfaces against corrosion and as catalysts. In the emerging field of nanotechnology, a goal is to make nanostructures or nanoarrays with special properties with respect to those of bulk or single particle species. Oxide nanoparticles can exhibit unique physical and chemical properties due to their limited size and a high density of corner or edge surface sites. Particle size is expected to influence three important groups of basic properties in any material. The first one comprises the structural characteristics, namely the lattice symmetry and cell parameters. Bulk oxides are usually robust and stable systems with well-defined crystallographic structures. However, the growing importance of surface free energy and stress with decreasing particle size must

be considered, changes in thermodynamic stability associated with size can induce modification of cell parameters and structural transformations and in extreme cases the nanoparticle can disappear due to interactions with its surrounding environment and a high surface free energy. In order to display mechanical or structural stability, a nanoparticle must have a low surface free energy. As a consequence of this requirement, phases that have a low stability in bulk materials can become very stable in nanostructures. This structural phenomenon has been detected in TiO_2 , VO_x , Al_2O_3 or MoO_x oxides⁸⁻¹⁰. Cobalt-based nanoparticles reside between the most promising materials for technological applications like information storage, magnetic fluids and catalysts. The low crystal anisotropy of cobalt also promotes their study them as a model system for the effects of size, shape, crystal structure and surface anisotropy on their macroscopic magnetic response. A variety of methods to the preparation of cobalt colloid dispersions has been reported. The most common are the reduction of a cobalt salt and the thermal decomposition of a cobalt carbonyl^{11, 12}.

2. PROPERTIES OF NANOMATERIALS

At the nanoscale dimensions, the material properties change significantly differing completely from their bulk counterparts. Nanomaterials display new phenomenon associated with the quantized effects and with the preponderance of surfaces and interfaces. The quantization effect arises in nanometer regime because the overall dimensions of objects are comparable to the characteristic wavelength of fundamental excitations in materials. As the size of material decreases, the percentage of surface atoms increases, thus increasing the reactivity and making them highly reactive catalysts, as surface atoms are the active centers for elementary catalytic processes. For example, iron nanoparticles of sizes 3 nm, 10 nm and 30 nm will have 50%, 10% and 5% of atoms on surface. Among the surface atoms, atoms sitting on the edges and corners are more reactive than those in planes. Also, the percentage of atoms at the edges and corners increases with decrease in the particle size and therefore, smaller metal particles are preferred for catalysis. An interesting feature of metal and semiconductor nanoparticles is their optical property. These nanomaterials exhibit interesting shape and size dependent optical properties due to quantum confinement. However, in a bulk crystal, the properties of material depend on chemical composition and not on size. Due to the decrease in size of a crystal towards nanometer regime, the electronic structure is altered from continuous electronic bands to discrete or quantized electronic

levels. Therefore, the continuous optical transitions between the electronic bands become discrete and the properties of nanomaterial become size dependent¹³. Metal nanoparticle (Au, Ag and Cu) dispersions exhibit colors due to the surface plasmon resonance (SPR) phenomenon, which is caused by the coherent oscillation of conduction band electrons when they interact with electromagnetic field. During SPR, polarization of the electrons with respect to the much heavier ionic core is induced by the electric field of an interacting light wave. This creates a net charge difference at nanoparticle surface, which acts as a restoring force. Thus, a dipolar oscillation of all the electrons with the same phase is created. The so observed color originates from the strong absorption by the metal nanoparticles when the frequency of electromagnetic field becomes resonant with the coherent oscillation of electron motion. The frequency and width of SPR depends on the metal nanoparticle size, shape, dielectric constant of the metal itself and the surrounding medium. Similarly, other important properties such as electronic properties, magnetic properties, melting point and catalytic properties of the nanomaterials depending on their shape, size, composition and surrounding medium have been studied in great detail¹⁴. Another most important property of nanomaterials, especially semiconductor nanoparticles, is to exhibit photocatalytic behavior. Size dependent properties (quantization) of semiconductors (such as TiO₂ and ZnO etc.) and quantized charging effects in metal nanoparticles provide the basis for developing new and effective systems. These nanostructures provide innovative strategies for designing next generation energy conversion devices. In a semiconductor, the energy difference between the valence band and the conduction band is known as the “Band Gap”. When it absorbs radiation from sunlight or illuminated light source, it generates pair of electron and hole. This stage is known as the semiconductors, photo-excitation state. The recombination of electron-hole pair may result in light emission with appropriate further reactions. These semiconductors behave as photo catalysts¹⁵. Nanotechnology has created a kind of revolution as this new area encompasses physics, chemistry, materials science and engineering and also biology and medicine. Several applications are envisaged from these interesting materials in the field of sensors, catalysis, diagnostic tools, therapeutic agents, drug/gene delivery vehicles, solar cells, plasmonics devices, cosmetics, coating materials, cell imaging, fuel cells, photonic band gap materials, single electron transistors, non-linear optics devices and surface enhanced Raman spectroscopy¹⁶. Two principal factors cause the properties of nanomaterials to differ significantly from other

materials: increased relative surface area and quantum effects. To understand the effect of particle size on surface area, consider a U.S. silver dollar. The silver dollar contains 26.96 grams of coin silver, has a diameter of about 40 mm and has a total surface area of approximately 27.70 square centimeters. If the same amount of coin silver were divided into tiny particles - say 1 nanometer in diameter, the total surface area of those particles would be 11,400 square meters. When the amount of coin silver contained in a silver dollar is rendered into 1 nm particles, the surface area of those particles is 4.115 million times greater than the surface area of the silver dollar!¹⁷.

Electrical properties: The electrical properties of nanomaterials vary between metallic to semiconducting materials. It depends on the diameter of the nanomaterials. The very high electrical conductivity of nanomaterial is due to minimum defects in the structure.

Thermal conductivity: The thermal conductivity of nanomaterials are very high, is due to the vibration of covalent bonds. Its thermal conductivity is 10 times greater than the metal. The very high thermal conductivity of nanomaterial is also due to minimum defects in the structure.

Mechanical properties: Nanomaterials are very strong and withstand extreme strain. Most of the materials fracture on bending because of the presence of more defects, but nanomaterials possess only few defects in the structure¹⁸.

3. CARBON NANOSTRUCTURE - PROPERTIES AND APPLICATIONS

Single walled carbon nanotubes (SWCNT) form a hexagonal network of carbon atoms rolled into a seamless, hollow cylinder, with each end capped with half a fullerene molecule. Rolling of the graphene sheet at different angles creates a visible twist or spiral in the molecular structure, giving it chiral properties. The basic structure of a SWCNT is specified by a single vector called the chiral vector Ch . As shown in Fig. 1, this vector is denoted by OA and is the section of the nanotube perpendicular to the nanotube axis, which when rolled up is the circumference of the tube. Fig. 1 shows the unrolled lattice of the nanotube and, in this case, the direction of the nanotube axis is given by the vector OB . The lattice can then be rolled to form a cylinder by lining up the points so that O is on top of A and B is on top of B' ¹⁹. Carbon nano tubes (CNT) can be found in other forms, for example the double walled carbon

nanotubes (DWCNT) (Fig. 2b) and multi walled carbon nanotubes (MWCNT) (Fig. 2a and 2c). DWCNT and MWCNT contain two and more coaxial cylinders, respectively, with each cylinder being a SWCNT with weak van der Waal forces binding the cylinders together.

Carbon nanofiber (CNF) is characterized by a graphitic like structure with variable alignments of laminated hexagon layers along the fiber axis. The graphitic platelet sheets that make up the nanofiber can be mainly divided into three different configurations, as shown in Fig. 3. Fig. 3b shows that the graphitic layers are at an angle to the fiber axis and this is usually called the herringbone or cup stacked structure due to its shape. In the final configuration, graphitic layers are positioned perpendicular to the fiber axis and are stacked on top of each other (Fig. 3c)²⁰. Due to their remarkable properties, CNTs have been investigated for various applications such as light emitting diodes, sensors, actuators, logic circuits, super capacitors, filters and ceramic and reinforced composites. CNFs are used in fuel cells, super capacitors sensors and tissue engineering and composite applications²¹.

Electrostatic force microscopy (EFM) has been widely used to investigate electrostatic properties of nanomaterials. In EFM experiments, an AC bias voltage with frequency ω is applied between a conducting atomic force microscope (AFM) probe and the sample. A lock-in amplifier at the same AC frequency ω is used to detect interaction forces between the tip and the electrostatic field due to static charges and permanent dipole moments of the sample. In addition, a dipole moment in the sample induced by the AC bias voltage also interacts with the tip. This dielectric force oscillates at the double frequency and is thus detected using a separate lock-in amplifier at the 2ω frequency. The electrostatic force channel in EFM has been utilized in investigating nanometer-scaled distribution of charge and electric dipole moment, surface potential variation, charge storage, and leakage in materials and devices. The 2ω channel in EFM has also been utilized to measure the equivalent dielectric constant of individual SWCNT²².

4. SYNTHESIS OF NANOMATERIALS

Nanomaterials can be synthesized by any one of the following methods

Pyrolysis: It involves pyrolysis of hydrocarbons such as acetylene at 7000C in the presence of Fe-silica or Fe-graphite catalyst under inert conditions.

Carbon arc method: It is carried out by applying direct current (60-100 A and 20-25 V) arc between graphite electrodes of 10-20 μ m diameter.

Laser evaporation method: It involves vapourisation of graphite containing small amount of Co and Ni, by exposing it to laser beam at 12000C in a quartz tube reactor. An inert gas like argon is allowed to pass into the reactor to sweep the evaporated carbon atoms from the furnace to the copper collector, on which the nanomaterials condense.

Chemical vapour deposition: It involves decomposition of vapour of hydrocarbons such as methane, ethylene, acetylene, etc., at 11000C in presence of catalysts like Ni, Co, Fe supported on MgO. Fig. 4 shows the schematic outlines of the various approaches for the synthesis of nanoparticles²³.

Combustion synthesis (CS) or self-propagating high-temperature synthesis (SHS) is an effective, low-cost method for production of various industrially useful materials. Today CS has become a very popular approach for preparation of nanomaterials and is practised in 65 countries. Recently, a number of important breakthroughs in this field have been made, notably for development of new catalysts and nanocarriers with properties better than those for similar traditional materials. The extensive research carried out in last five years emphasized the SHS capabilities for materials improvement, energy saving and environmental protection. The importance of industrialization of the SHS process is also realized. All these aspects were adequately brought out and discussed in the international conference devoted to the 40th anniversary of SHS, which was held at ISMAN (Chernogolovka, Russia) in October 2007²⁴. Nanostructured ZnO materials have drawn broad attention due to its wide range of applications in ultraviolet (UV) lasers, power generators, solar cells, gas sensors, field emission devices, capacitors, varistors, transparent UV resistance coating, photoprinting, electrophotography, electrochemical and electromechanical nanodevices, sun screen lotion (cream), cosmetic and medicated creams etc. Some of the future applications include catalysts for organic reactions and delivering drugs to infected areas of body.

Many synthetic methodologies for zero and one dimensional ZnO nanostructures have been developed, including vapor phase methods and aqueous methods. The vapor-phase method usually employs vacuum, sophisticated equipment and elevated temperature which restrict the type of

substrate used, in contrast to the low-cost and simple aqueous methods. However, simple methods for preparation of high quality nanostructures with high yield and good control on shape and size are very less. Among the aqueous methods, the hydrothermal method has emerged as a powerful method for the fabrication of one-dimensional nanomaterials with some significant advantages, such as controllable structures and a cost effective, low-temperature, substrate independent and less complicated technique. However, yield and quality of the product material is usually not addressed. For example, Kale et al. reported synthesis of high-yield of flowerlike ZnO nanorods using a hydrothermal method²⁵. However, near band edge UV emission was not observed from those samples due to poor quality of the material and large defect concentration. Similarly, large-scale synthesis of ZnO flower-like and brush pen-like nanostructures by a hydrothermal decomposition route showed very weak UV emission and strong green emission²⁶. In majority of the studies, the product suffers from poor optical properties due to presence of large concentration of intrinsic defects in ZnO. Chu et al recently reported a solution-based, high yield synthesis of cobalt-doped ZnO nanorods with roomtemperature ferromagnetism²⁷. However, the reported studies have product yield below 50%. Hence, large scale and high yield synthesis of defect free ZnO nanostructures is imperative to meet the demand for emerging applications²⁸. Cuprous iodide (CuI) has attracted much attention because of its unusual features such as large band gap, a negative spin-orbit splitting, an unusually large temperature dependency, anomalous diamagnetism behavior, large ionicity, new high pressure phase etc. and potential applications in superionic conductor, solid-state solar cells, catalysis for synthesis of organic compounds and others. Nano size CuI is being pursued with great interest because of several possible technical applications in catalysis, drug delivery systems, separation techniques, photonics as well as piezoelectric and other dielectric devices²⁹. Fluorescent nanoparticles have attracted increasing research attention due to their promising applications covering electrooptics to bionanotechnology. To date, typical photo luminescent particles have been developed from compounds of lead, cadmium and silicon. But these materials also have raised concerns over potential toxicity, environmental harm and poor photostability. The need for photoluminescent nanostructures emitting in visible-to-near infrared (NIR) spectral range is rapidly increasing. Compared to conventional measurements made in the ultraviolet visible region, spectrofluorimetry within the therapeutic window of

700-1200 nm has many advantages, such as lower levels of background interference and deeper penetration of living tissues. NIR photoluminescence (PL) obtained from NIR excitation holds great promise for in vivo uses at significant depths in biological media and the development of noninvasive diagnostic techniques. Consequently, an excellent fluorescent nanoparticle for bio applications should be more stable, and less toxic as well as exhibit good NIR emission. Compared to traditional quantum dots (QDs) and organic dyes, photo luminescent carbon nanomaterials are superior in chemical inertness and likely lower toxicity. The emergence of photo luminescent carbon-based nanomaterials has presented exciting opportunities for searching for fluorescent nanomaterials. Moreover, the up-conversion nanomaterials for biological luminescent labels also have attracted great attentions due to their unique luminescent properties with long lifetime and superior photo stability. In comparison with down conversion fluorescent materials, up-conversion materials have many advantages in biological applications, such as noninvasive and deep penetration of NIR radiation, the absence of auto fluorescence of biological tissues, and feasibility of multiple labeling with different emissions under the same excitation. A recent study indicated that core-shell structured CNPs have two-photon active luminescence with excitation in the NIR range. Thus, we can expect that carbon-based fluorescent nanoparticles may possess visible, NIR, and up-conversion luminescence synchronously, which are extremely important for fundamental and practical applications³⁰.

5. NANOCRYSTALS

Nanocrystals are zero-dimensional particles and can be prepared by several chemical methods, typical of them being reduction of salts, solvothermal synthesis and the decomposition of molecular precursors, of which the first is the most common method used in the case of metal nanocrystals. Metal oxide nanocrystals are generally prepared by the decomposition of precursor compounds such as metal acetates, acetylacetonates and cupferronates in appropriate solvents, often under solvothermal conditions. Metal chalcogenide or pnictide nanocrystals are obtained by the reaction of metal salts with a chalcogen or pnictogen source or the decomposition of single source precursors under solvothermal or thermolysis conditions. Addition of suitable capping agents such as long-chain alkane thiols, alkyl amines and trioctylphosphine oxide (TOPO) during the synthesis of nanocrystals enables the control of size and shape. Monodisperse nanocrystals are obtained by post-synthesis size-

selective precipitation³¹. Solid metallic nanoparticles are well-known for their attractive optical properties, strong optical resonance and a large and fast nonlinear optical polarizability associated with the Plasmon frequency of the conduction electron in the particle. Colloidal metal shells on the other hand can have resonance that can be tuned over a wide range as a function of the core-to-shell ratio. The greatly enhanced ability to manipulate the boundary conditions of the resonating conduction electrons makes it possible to cover theoretically the ultraviolet, visible and infrared parts of the spectrum. Westcott et al. have shown in several papers how such shells composed of gold can be grown around silica colloids and how the single particle properties can be exploited. Silica colloids provide for a convenient dielectric core as they can be grown with a small polydispersity³².

6. EXAMPLES OF WHERE NANOTECHNOLOGY IS BECOMING USED

Nanowire and nanotube arrays for EMI shielding: Composite materials with superior thermal, electrical and mechanical properties, metal and semiconductor nanowires as tags for bioassays, etc. Carbon nanotubes, seamless tubes of graphite sheets of nanosized diameter, include SWCNT and MWCNT. In some cases, the ends of the nanotubes are open, whereas in other cases they are closed off with full fullerene caps. Depending on sheet direction and diameter a nanotube may be either metallic or semi-conducting. Compared with all other kinds of natural materials, carbon nanotubes have the highest theoretical strength, although their specific gravity is only one-sixth of steel. Carbon nanotubes provide special advantages in shielding and absorbing electromagnetic radiation, field emission, thermal conductivity, hydrogen storage, adsorption, catalyzing, etc.

Chemical gas sensing: Robust low power micro sensors and micro sensor arrays with high sensitivity and selectivity, such as humidity sensors, solid state resistive sensors, combustible gas sensors, etc.

Ceramic MEMS: 2D and 3D micro components and micro electromechanical devices for harsh application environments, high aspect ratio MEMS, micro channel plates, etc.

Energy conversion: Photo-voltaics, radiation detection, electroluminescent and lasing materials and devices, components for mesoscopic energy sources, etc.

Electronics and related fields: Scanning probes and scanning microscopy standards, storage media and Terabit memory, flat panel displays, vacuum microelectronics for harsh environments, field emission cathodes, photonic band gap materials and devices, etc.

Marine anti-fouling: Due to the small size of the nanomaterials, the particles are held in the coating lattice and are not readily leached out of the coating by the marine environment, while slowly releasing ions to help provide longer-term anti-fouling character. The need for longevity of antimicrobial activity in marine environments is widely recognized.

Semiconductor polishing: As the semiconductor industry continues to move forward to smaller chip architecture, the need for advanced CMP slurries becomes a requirement that cannot be met by the slurries provided in the past. Currently fumed silica and colloidal silica are being used. Both of these are nanoparticles used for several decades already. New types are being developed now including ceria, mixed rare earth metal oxides and alumina dispersions which are on the forefront of providing high planarity surfaces and efficient removal rates. The unique surface chemistry of these nanomaterials allows formulation of highly concentrated dispersions at a variety of pH. Cabot is another leader developing products for this market.

Sunscreen formulations: Nanomaterials can act as sun blockers to protect human skin in formulations that go on smooth, silky and clear. Eliminating unnecessary exposure to the harmful UV rays of the sun has increasingly become a key health concern.

Catalysts: The use of nanomaterials based on rare earth metal oxides allows for the preparation of thinner active layers, which can mean less precious metal usage. These nanomaterials also allow for the preparation of higher solids dispersions that are very stable, minimizing the number of coating steps and losses due to flocculated dispersions. Automotive catalytic converters are a key focus area for catalyst performance. One way to achieve lower emissions in a cost effective manner is to utilize co-catalysts that provide good oxygen storage capability and thermal stability in thinner layers. Nan ceria and mixed rare earth metal oxides meet the criteria necessary to enhance catalytic converter performance when properly incorporated into a catalyst system, and because they are dense, single phase individual crystals, there is nothing to collapse during thermal cycling³³.

7. CONCLUSIONS

Nanoscience refers to the science & discipline and nano technology refers the applied part of it including the engineering to control, manipulate and structure the matter at an unimaginably small scale: nano scale. Nanotechnology is one of the most active research areas with both novel science and useful applications that has gradually established it in the past two decades. Expenditure on nanotechnology research is significant; however, the research is continuously

moving forward motivated by immediate profitable return generated by high value commercial products. The mankind has seen several technology revolutions in the past: industrial, agricultural, medical and infotech in a course of two centuries. Chemical sensors from nanoparticles and nanowires enhanced the sensitivity and sensor selectivity. The main aim is to enhance productivity through nano tech driven precision farming and to maximize the output and minimize inputs through better monitoring and targeted.

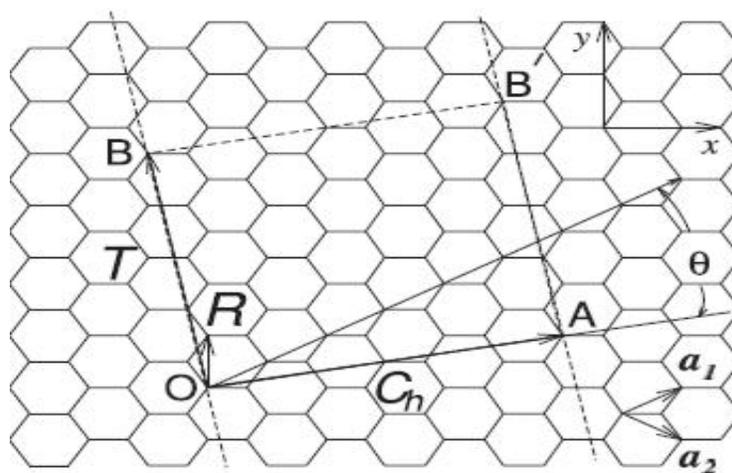


Fig. 1: Chirality in nanotubes

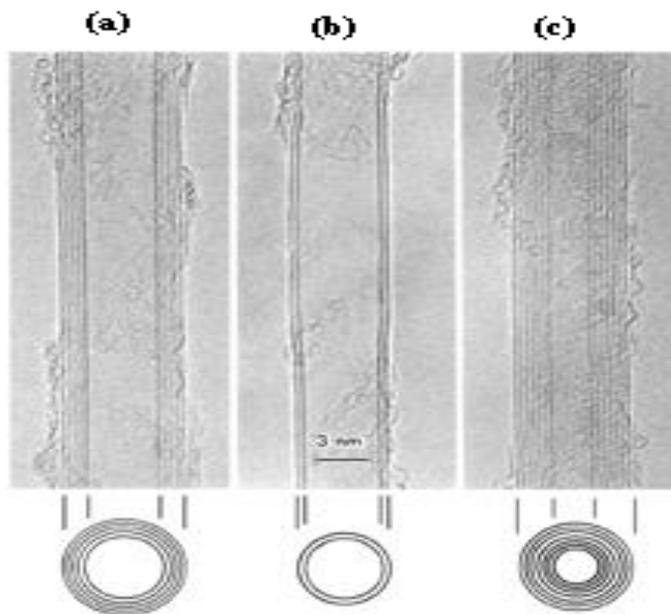


Fig. 2:(a) MWCNT (b) DWCNT and (c) MWCNT

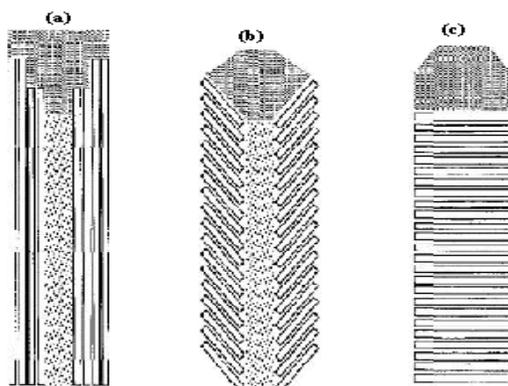


Fig. 3: Schematic representation of various arrangements of graphitic platelets in carbon nanofibers (a) tubular (b) at angle and (c) perpendicular to the fiber axis

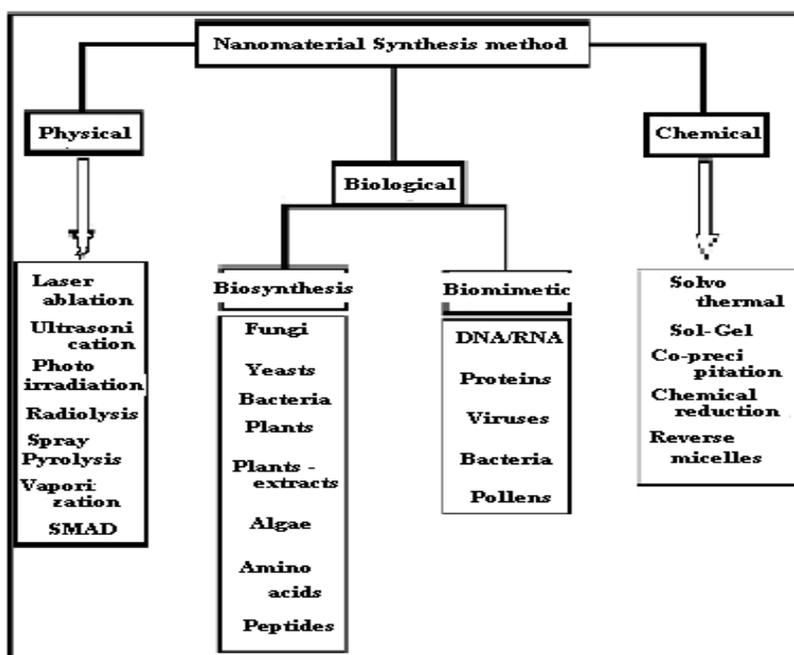


Fig. 4: Schematic outlines of the various approaches for the synthesis of nanoparticles

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