Chapter 9

Biomolecules and Pure Carbon Aggregates: An Application Towards "Green Electronics"

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Additional information is available at the end of the chapter

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Abstract

"Green electronics" is a novel scientific term which aims to identify the compounds of natural origin (economically safe and biodegradable) and establish economically efficient route for production of synthetic materials. The purpose of green electronics is to create path for the production of human and environmental friendly electronics and the integration of electronics with living tissue in particular. These researches may help to fulfill not only the organic electronics to deliver low cost energy efficient materials and devices, but also achieve unimaginable functionalities for electronics. In this chapter we have considered the molecular electronic devices biomolecules: deoxyribonucleic acid (DNA) and pure carbon aggregates: (carbon nanotubes (CNTs)/graphene), their properties and applications.

Keywords: biosensing, carbon nanotubes (CNTs), graphene, nucleobases, sensors

1. Introduction

Nanobiotechnology is gaining tremendous impetus in this era due to its ability to modulate metals into their nanosize and further interaction to the biological complexes, which efficiently changes their physicochemical and optical properties. Accordingly, considerable attention is being given to the development of novel strategies for the different nanoparticles of specific composition and size using biological sources. As the currently available techniques are expensive, environmentally harmful, and inefficient with respect to materials and energy use, so the emphasis is given to design the user friendly, non-toxic complexes, which can be used in biomedical and environmental applications. The major key prerequisite for achieving sustainability in the electronics industry is the usage of materials and technologies that have low embodied energy. Numerous efforts have been made throughout the world to develop environmentally benign technologies with nontoxic products using green nanotechnology and biotechnological tools. The synthesis of these nanoparticles using biological methods or green technology has diverse nature, greater stability and appropriate dimensions. So it is the current demand of the entire world to use specific techniques to characterize the potential of these hybrid complexes as an application towards drug delivery and biomedical fields. As we are focused towards Molecular electronics, so *bottom up* approach is included in this chapter, in which the individual molecular devices are built by synthesizing molecules with the desired electronic properties which are interconnected into an electronic circuits using attachment techniques like self-assembly. The main advantages of these complexes are their natural nanoscale structures, which can be created as absolutely identical in vast quantities and low fabrication cost [1–8] (**Figure 1**).

The reason for green electronics arises as the elements used previously in the field of green electronics such as Pb, Cd, Zn, Cu, Cr and As are potential bio-accumulative toxins in the production system of milk and dairy products [9]. Cadmium cause bone demineralization, either through direct bone damage or indirectly as a result of renal dysfunction [10].

In contrast to the above, the application of nanoscale materials for electrochemical biosensors has been grown exponentially due to high sensitivity and fast response time [11, 12]. So we can say that there evolved a parallel study as nano-bio-complexes in the field of green electronics, blending the biomolecules and nanotechnology. These nano-biosensor designs have created revolution in the field which can become a pioneering research work. Recently hierarchical cluster analysis (HCA) and principal component analysis (PCA) [13] were used to assay the receptor signaling mechanisms which can be used in biosensors or nano-biosensors. The findings above lead to amperometric biosensor [14] based on enzyme from *Brassica napus* hairy roots to determine ochratoxin, is a colorless crystalline compound that is classified as pentaketides [15]. All biological molecules and cell organelles are chemo mechanically controlled systems known to every biologist. It is an interdisciplinary art to activate them to work as an electronic device [16]. Above all, developing an Immunosensor depends on immobilization of

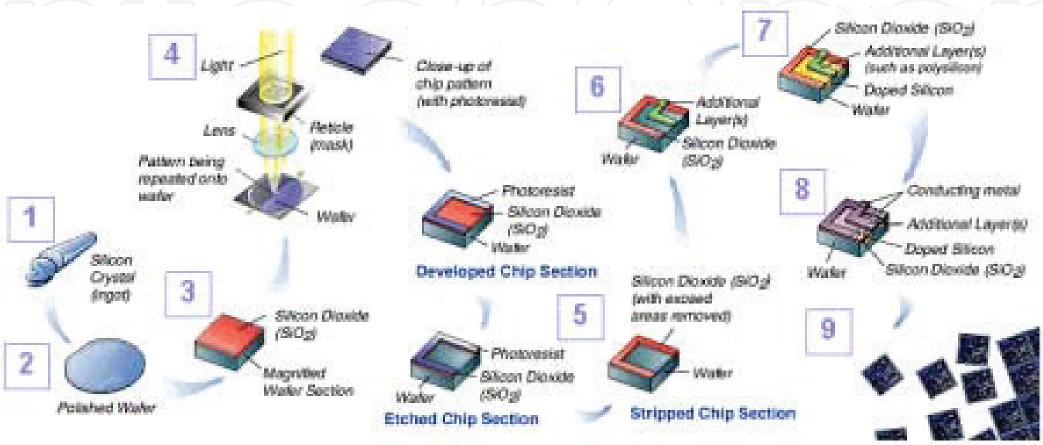

Figure 1. Schematic sequence of processes in the construction of a commercial chip (image from official INTEL website http://www.intel.com).

antibody molecules, which becomes an important factor for successful fabrication of immunosensors [17], which involves screen printing technology. Photoacoustic imaging technique has been developed which works on ultrasound spatial resolution and intrinsic rich optical contrast, penetrates deep into the tissues [18, 19] with acoustic ultra sounds and act as a detection tool in diagnostic medicine [20]. Thomas et al. [21], demonstrated all aspects of overhand throwing, using a 12 camera Vicon motion analysis system in a general motor program using bio-electronic signals. The synthesis from bioprocess or the material used as biomaterial, or the organism used for "GE" i.e. "Genetics Engineering," or the sensor and the receptor in the field, there is a relation between each research studies; that is nothing but the *field of biotechnology*.

Molecular electronics (ME) deals intensively with a long term alternative for increasing the device density in an IC and continuing Moore's law down to the nanometer scale. The basic idea of ME is to use individual molecules to act as wires, switches and memories. Till date, no detailed investigations have been carried out for various complexes using green technology, their utilization, or their analyses have been published. Therefore, this chapter is conducted to highlight the use of various nano-bio complexes, their use in green technology and different techniques for characterization of nanoparticles to provide a better understanding of the these sources to improve their uses in modern technology.

The ME can be further divided to (a) small covalently bonded organic molecules ((aromatic chains, conjugated polymers); b) large biomolecules (DNA, nucleoside-based aggregates, proteins); c) pure carbon aggregates (carbon nanotubes, graphenes).

The organic electronics based on conjugated polymers or small molecules as the core semiconductor element hold the high promise of delivering low-cost and energy-efficient materials and devices, yet the performance and stability of organic semiconductors remain major hurdles in their development as solid competitors of the inorganic counterparts. So the "soft" nature of carbon-based materials are considered enabling fabrication of extremely flexible, highly conformable and even imperceptibly thin electronic devices. In recent years, the graphene based nanomaterials have received considerable attention owing to their distinguished electronic and transport properties and act as promising candidates for electronics and spintronics. So green materials can act as emerging concept with carbon based class and integration of electronics into living tissue with the aim of achieving biochemical monitoring, diagnostic drug delivery tasks or generating human and environmentally benign technologies. Now the green technologies are carving the avenues towards achieving the ambitious goal of sustainability in the field of electronics. The quest is to achieve electronics sustainability by solving the energy deficiency puzzle and redressing the unfolding disaster, for which we look to the apparent simplicity of nature. Our aim is to create a novel class of engineered materials which are able to deliver complex functions that found applications in electronics; designing superhydrophobic (lotus effect), super-adhesive (gecko effect) or self-healing surfaces. Nature is the most efficient energy consumption engine that can be used for infinite purposes. In the last 10 years, we have witnessed a great deal of effort towards the development of novel conductive materials (electrodes) able to interface electronics with biological matter to deliver recognize events (i.e. biosensing, bio-recognition) and the modulate events (i.e. tissue engineering). Here we have selected the larger biomolecules and pure carbon aggregates and their role in the field of green electronics.

2. DNA biomolecular electronics

The role of ME is to provide reproducible well-structured architectures, easy to wire in a programmed manner. Supramolecular chemistry seems to fulfill these needs [22]. The two properties which are attractive for this purpose is the (a) molecular recognition and (b) self-assembly. Molecular recognition is the capability of a molecule to form selective bonds with other molecules or with substrates, which rest on the information stored in the structural features of the interacting partners. Molecular recognition processes (a) the building up of the devices from their components (b) incorporate them into supramolecular arrays; (c) allow selective operations on given species (e.g. ions, dopants), and (d) control the response to external perturbations (e.g. external fields, light, electrons, other molecules, etc.).

The self-assembly has the capability of molecules to spontaneously organize in supramolecular aggregates under well-defined experimental conditions. Self-organization may occur both in solution and in the solid state, and make use of hydrogen bonding, electrostatic donor-acceptor effects (Van der Waals, dipolar, etc.) or metal-ion coordination as basic interactions between the components. Due to these two properties, DNA molecules seem particularly suitable to be used as components for the construction of nanometer scale devices [23–26]. The idea of using DNA in molecular devices is its natural function of storing and coding the genetic information. DNA transmits well-defined chemical information through the pairing properties of the bases. In addition, it occurs in a large variety of structures and display physiochemical stability and mechanical rigidity.

2.1. Electron transfer through DNA

The deoxyribonucleic acid (DNA) is a biopolymer in a double helical form, which is constituted by an extended array of aromatic π-stacked base pairs adenine-thymine (AT) and guanine-cytosine (GC) within a polyanionic sugar-phosphate backbone (**Figure 2**). Due to the biological implications, the studies about the charge migration in DNA were related to physiological processes: the possibility and efficiency of charge transfer is significant, because the migration of the radical cation is a critical issue to understand problems related to radiation damage and mutation [27, 28].

The role of π-π interactions between stacked base pairs in double-stranded DNA could provide a pathway for rapid one-dimensional charge separation. Various experiments were performed

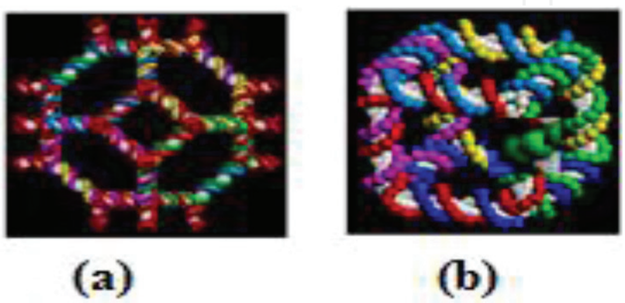

Figure 2. Representations of left (a) truncated octahedron containing six squares and eight hexagons. (b) Each edge of the truncated octahedron contains two double helical turns of DNA a DNA cube containing six different cyclic strands. Their backbones are shown with different colors.

to understand whether DNA facilitates the charge transfer over long distances and whether the base pair stack can act as a conducting medium. The issue of charge migration in DNA has recently become a hot topic with solution chemistry (in particular) after the first reports by Jacqueline Barton's group [29–33] in the early '90s. Although the answer to the question: Is DNA a molecular wire? is still elusive.

The recent achievement is the construction of few DNA-hybrid devices, which requires the application of some state-of-the-art nanotechnologies are: electron beam lithography for the fabrication of metallic nanocontacts, trapping techniques to compel the molecules into the desired device scheme, Atomic Force Microscopy (AFM) or Scanning Tunneling Microscopy (STM) for imaging and probing samples.

The achievement of DNA-based devices requires [33]:

- Construction of nucleic-acid networks.
- Conversion of nucleic-acid (or DNA-protein) network into electron conducting system.

The DNA-based materials may be used either as conductive wires or as a template for other conductive materials. By exploiting the molecular recognition of its functional groups, it is possible to synthetize branched DNA-motifs that may be assembled into periodic arrays. Though a lot of research conducting in this field is giving the contrasting results.

The most important ones are:

1. The intrinsic properties of the different DNA molecules employed in the experiments, the length of the DNA (from a few nanometers to some microns) and the structural conformation of the double helix.
2. The properties of the buffer solution in which the DNA is kept and the presence and the concentration of counter ions;
3. The experimental conditions in which measurements are realized: in air, in vacuum, different humidity, and different temperatures (from 1 to 300 K);
4. The structural aggregation forms of DNA (films, network bundles, single molecules);
5. The presence of contacts and the effects of the DNA/electrode junction.

Apart from experimental difficulties in the fabrication of DNA-based device, several fundamental questions are still open: what are the interactions which control the electrical properties of DNA? How do they depend upon the sequence? What are the mechanisms for charge transport? What are the effects of dopants or defects? How does DNA attach to a metal electrode? What are the effects of the contacts on the conduction properties of the device?

2.2. Application of DNA in green electronics

DNA can be used in various applications in green electronics, which is discussed herein.

2.2.1. G4-wires DNA as nanowire

Nanowires known as G4-wires [34] (or quadruplexes), consist of stacked guanine (G) tetrads (G4). These one-dimensional polymers act as prospective candidates for bio-molecular electronics because, due to the low ionization potential of guanine (the lowest among nucleic-acid bases), they might be suitable to mediate charge transport by hole conduction along the helix, and have even been suggested as nano-mechanical extension-contraction machines [35]. In the presence of appropriate metal cations (especially K^+ and Na^+), solutions of homo-guanylic strands in water [36, 37] as well as lipophilic guanosine monomers in organic solvents [38], self-assemble in right-handed quadruple helices. Recent investigations has been carried out for the G4-nanowires, but the conduction properties of these nanowires are basically unknown and a direct measurement of electrical properties of G4-wires is still missing.

2.2.2. DNA as sensors and imaging agents for metal ions

Sensing and imaging of metal ions have attracted much attention by scientists and engineers because of the important roles of metals in many fields such as environmental, biological, and medical sciences. Significant progress has been made in developing sensors and imaging agents for the detection of metal ions, mostly based on organic molecules, peptides, proteins, or cells [39–45]. Prof. Yi Li has given a significant insight to the role of DNA as sensors and imaging agents for metal Ions [46], the structure of these systems is illustrated and described in **Figure 3**. DNA does not appear to be a good candidate for sensing metal ions with high selectivity because the negatively charged phosphodiester backbones of DNA are known to be capable of binding cationic metal ions with poor selectivity for any particular metal ion. While the four DNA bases can also serve as ligands for metal ions [47–49], many of these DNA–metal ion interactions are nonspecific and weak, making the use of DNA as sensors for metal ions very challenging because selectivity and sensitivity are required for the successful detection of a specific metal ion in the presence of other potentially interfering metals in

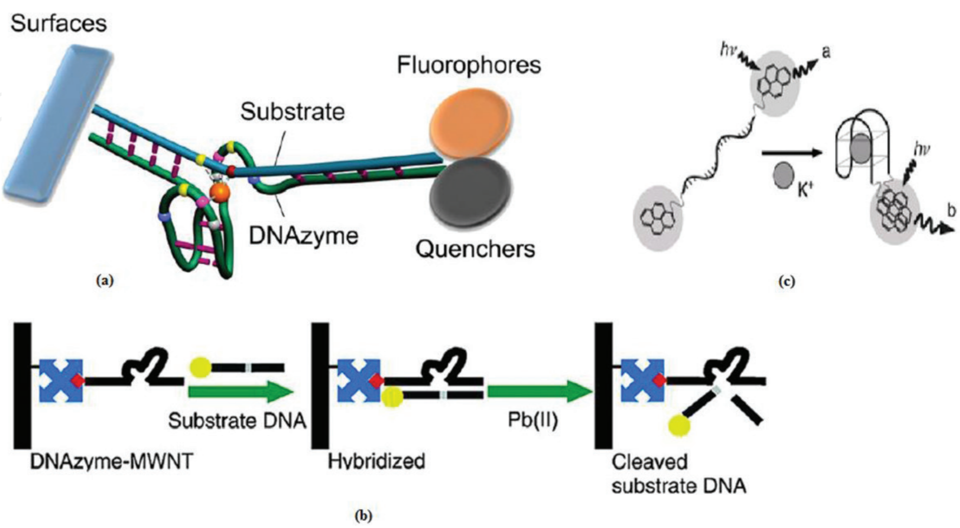

Figure 3. a. General sensor design based on nucleic acid cleavage of DNAzymes for metal-ion detection. b. Fluorescent Ag^+ sensor based on C–Ag^+–C. c. Sensors based on G-quadruplex DNA stabilized by K^+. Adapted with permission from Ref. [46].

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complex samples. By incorporation of signal reporters such as chromophores, fluorophores, electrochemical tags, and Raman tags, these metal-ion-specific DNA sequences have been transformed into colorimetric, fluorescent, electrochemical, and Raman sensors and imaging agents for a broad range of metal ions with high sensitivity and selectivity [50–55]. DNAzymes that is highly selective to use specific metal ions as cofactors to catalyze reactions can be obtained. In this way, DNAzymes that are dependent on bivalent metals for various chemical and biological reactions have been successfully discovered. One report of DNAzyme sensor was a fluorescent sensor for Pb^{2+} based on DNAzyme [56–58], which showed much higher specificity to Pb^{2+} over other metal ions in catalyzing the cleavage of DNA substrates with a single RNA linkage (rA) at the cleavage site.

These sensors can be further classified into different parts:

1. Fluorescent sensors based on metal ion-dependent DNAzymes
2. Fluorescent sensors labeled with fluorophores and quenchers [59]
3. Surface-immobilized fluorescent sensors [59]
4. Label-free fluorescent sensors [60–62]
5. Colorimetric sensors based on metal ion-dependent DNAzymes
6. Colorimetric sensors based on gold nanoparticles [63, 64]
7. Colorimetric "dipstick" tests using lateral-flow devices [65, 66]
8. Electrochemical and Raman sensors based on metal ion-dependent DNAzymes [67]
9. Sensors based on metal binding structures [68, 69]
10. Hg^{2+} sensors based on T–Hg^{2+}–T-containing DNA
11. Ag^{+} sensors based on C–Ag^{+}–C-containing DNA
12. Sensors for K^{+}, Pb^{2+}, Cu^{2+}, and Ag^{+} based on G-quadruplex DNA
13. Combination based sensors (DNAzymes and metal-binding DNA structure) [70]
14. Portable sensors [71]

In this category, new technologies have been developed to design sensors based on commercialized devices, compatible with portable devices, which could enable to monitor metal ions by all.

2.2.3. DNA-electrochemical biosensors

DNA biosensors are the integrated receptor-transducer devices that use DNA as biomolecular recognition element to measure specific binding processes with DNA, by electrical, thermal or optical signal transduction methods. The characteristics of DNA probes with the capacity of direct and label-free electrochemical detection find applications in rapid monitoring of pollutant agents or metals in the environment, investigation and evaluation of DNA-drug interaction mechanisms, detection of DNA base damage in clinical diagnosis, or detection of specific DNA sequences in human, viral and bacterial nucleic acids [72–75].

2.2.4. DNA oxidative biomarker

Oxidative DNA damage caused by oxygen-free radicals lead to multiple modifications in DNA, including base-free sites and oxidized bases. The damage caused to DNA bases is potentially mutagenic [76] and can be enzymatically repaired. The interest lies in the sensitive determination and full characterization of the mechanism involved in oxidative damage to DNA bases. Electrochemical methods are used to study the DNA oxidative damage and in the investigation of the mechanisms of DNA-drug interactions. In recent study, it has been anticipated that the mispairs-coinage metal complexes can also be used as a biomarker [77].

2.2.5. Electrochemical biosensors for detection of DNA damage

The DNA-electrochemical biosensor enables pre-concentration of the hazard compounds which are investigated onto the sensor surface and *in situ* electrochemical generation of radical intermediates, which cause damage to the DNA immobilized on the electrode surface and can be electrochemically detected.

2.2.6. DNA as radiation sensors

DNA has an optical response towards temperature, magnetic field, radiation and others. The flexibility of DNA can be modified by the radiations. When irradiated using gamma rays and neutrons (non-ionizing radiation), the dynamics of DNA macromolecules [78] changes its configuration when involved in environmental interactions with other components of the living cells [79]. Whenever any radiation passes through a semiconductor device, different effects are observed which depends on the range of energy of the particle (proton, alpha, neutron and both types of beta) and rays, such as gamma radiation [80]. These include defects as: vacancies, defect clusters, dislocation loops near the surface and adjustment of band gaps [81]. Electrical properties of DNA molecules can be understood by the electrical conduction mechanism, namely: thermionic emission, tunneling and hopping [82]. These all properties can be applied for DNA as radiation sensors.

3. Pure carbon aggregates

Carbon Nanotubes (CNTs) has interesting physicochemical properties as electrical conductance, high mechanical stiffness, light weight, transistor behavior, piezoresistance, thermal conductivity, luminescence, electrochemical bond expansion as well as their versatile chemistry make them superb materials for a broad spectrum of applications ranging from energy storage devices, nanosensors and drug/gene delivery vehicles.

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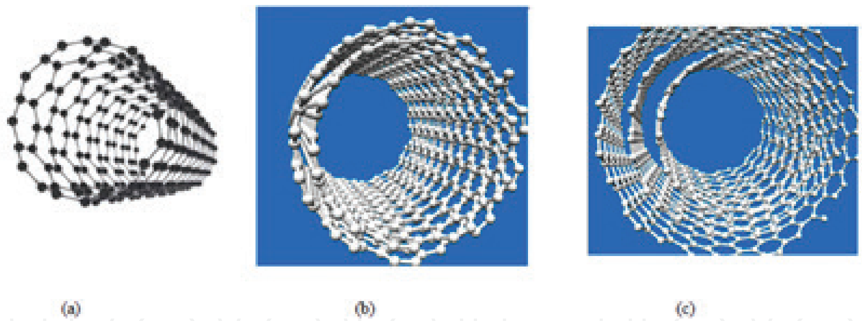

Figure 4. Representation of SWCNT and MWCNTs (armchair, zigzag and chiral).

3.1. Carbon nanotubes (CNTs)

Carbon nanotubes (CNTs) are allotropes of carbon with a cylindrical nanostructure, a member of the fullerene structural family. Due to their extraordinary thermal conductivity, mechanical, and electrical properties, carbon nanotubes find applications as additives to various structural materials. The various representations of CNTs are given in **Figure 4**.

3.1.1. Applications of CNTs in green electronics

Single-walled carbon nanotubes (SWCNTs) also have unique properties which make them suitable for applications in a variety of imaging modalities, such as magnetic resonance, near-infrared fluorescence, Raman spectroscopy, photoacoustic tomography, and radionuclide-based imaging.

The various features of carbon nanotubes are:

1. Drug delivery and cancer treatment [83] (**Figure 5**)
2. Physical cancer therapies delivered by CNTs [84]
3. Biosensing
4. Optical [85]
5. Electronic and electrochemical sensors [86]
6. Biomedical imaging
7. Molecular imaging [87, 88]
8. MRI with SWCNTs [89, 90]
9. Optical imaging with SWCNTs [91, 92]
10. Raman spectroscopy with SWCNTs [93, 94]
11. Photoacoustic tomography [95, 96]

12. Radionuclide-based imaging with SWCNTs [97, 98]

13. Scaffolds in tissue engineering

14. CNTs used as scaffolds in bone regeneration [99]

15. CNTs for neural applications

3.2. Graphene

The two-dimensional carbon material graphene find application for sensors, electronics and catalysis applications due to its exceptional electrical and mechanical properties. Some of these applications require the adsorption of metal clusters onto graphene and metal-graphene systems which are now become a subject of intense investigation. It show many interesting properties as the observable quantum Hall effect at room temperature [100, 101], existence of two-dimensional gas of massless Dirac fermions [102], ballistic transport properties on the sub micrometer scale [103], etc. As graphene is unique in nature, so researchers explore its unique properties in storage [104], spintronics [105], microelectronics [106], etc. A number of theoretical and experimental work have been carried out for the electronic and magnetic behaviors of dimers [107] and adatoms of different elements [108] adsorbed on graphene system, which have been found to yield many interesting results. Graphene also has immense potential to act as a key ingredient for new devices as single molecule gas sensors, ballistic transistors, and spintronic devices. Bilayer graphene, which consists of two stacked monolayers, has a quadratic low-energy band structure which generates very different scattering properties from those of the monolayer. It also presents the unique

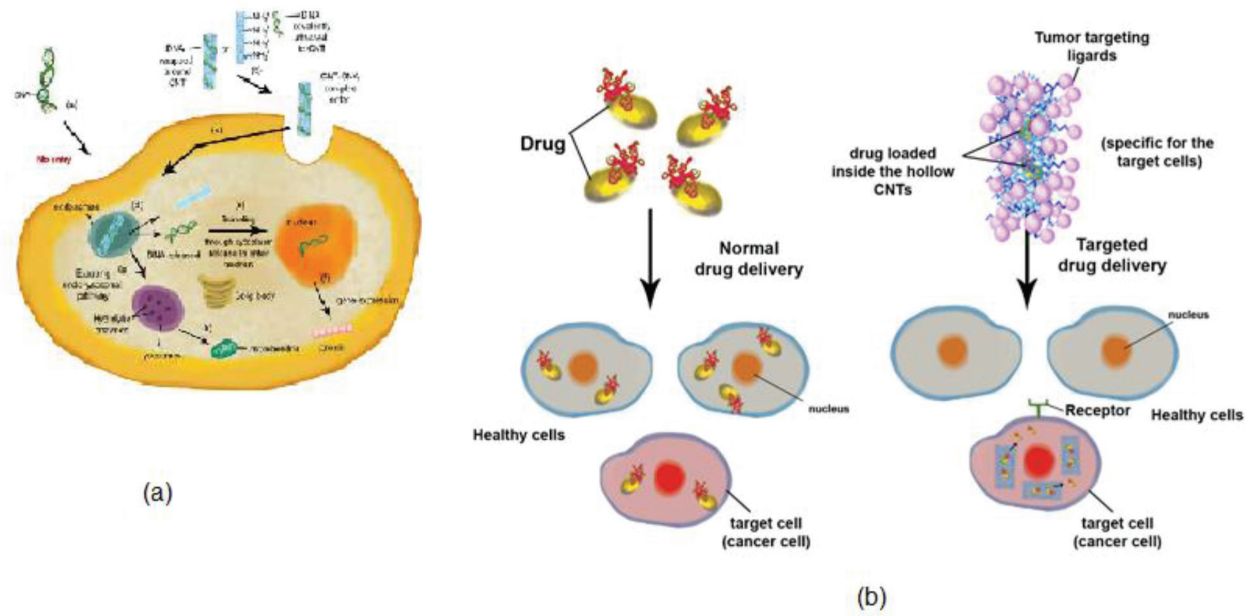

Figure 5. Representation of the role of CNTs in drug delivery. (a) Gene delivery by CNTs, (b) Normal drug delivery versus the efficient target drug delivery using CNTs.

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property i.e. *the tunable band gap can be opened and controlled easily by a top gate*. Another property of graphene is the high electronic mobility, which is crucial for many of its potential applications [109]. So, understanding the mechanism, which limit the mobility of carriers in graphene is extremely important. It is also of a high conceptual interest, since transport properties of chiral massless fermions are essentially different from those of conventional charge carriers in metals and semiconductors [110]. Charged impurity scattering has received the most attention [102], with the majority of studies modeling the impurities as point-like objects (1/r potential). Recently, it has been revealed in a theoretical study that the physical structure of the charged impurities and clusterization of charged impurities might be the an important factor, which influence their scattering properties [111]. Graphene nanoribbons (GNRs) present reactive edges which make GNRs not only more accessible to doping and chemical modification, but also more susceptible to structural defects [112]. In particular, for zigzag graphene nanoribbons (ZGNRs) terminated with one hydrogen atom on each zigzag edge, there are quite a few localized edge states near the Fermi energy level on both edges. Such localized edge states can lead to a spin induced energy gap [113] providing a significant effect on the electronic and transport properties [114]. The electronic and transport properties are thus very sensitive to the atomic structures and chemical modification of the edges. One of the natural ways of chemically modifying graphene is to include metal adatoms [115–117]. Motivated by the special transport properties of atomic wires of Al, Ag, Au and Cu, and inspired by the localized edge states of ZGNRs that greatly enhance the binding energy of adatoms, we also conducted the studies on (Al, Au and Cu) adatomed-objects on edge chlorinated nano graphenes to investigate the electronic, magnetic and adsorption and transport properties of these (C1 ($C_{42}CI_{18}$)/C2($C_{48}CI_{18}$)/C3($C_{60}CI_{22}$))–metal systems (**Figure 6**). Metals adsorbed on nanoscale carbon surfaces have been reported experimentally and theoretically to form a variety of structures, such as continuous coatings or discrete clusters [118] and novel interesting phenomena were observed to occur through suitable modification.

Metallic nanowires (namely, linear chains of metal atoms) have drawn significant attention due to the quantum confinement effect, as they represent the ultimate miniaturization of conductors. The nanoscales of metallic nanowires result in a number of novel and interesting phenomena different from their bulk materials [119–121]. Graphene based biosensors can be classified as follows:

1. Graphene-based electrochemical biosensors [122]
2. Graphene-based enzymatic electrochemical biosensors
3. Graphene-based bioaffinity electrochemical biosensors
4. Graphene-based DNA electrochemical sensors [123]
5. Graphene-based electrochemical immunosensors [124]
6. Graphene-based field-effect transistor (FET) biosensors [125]
7. Graphene-based optical biosensors [126]

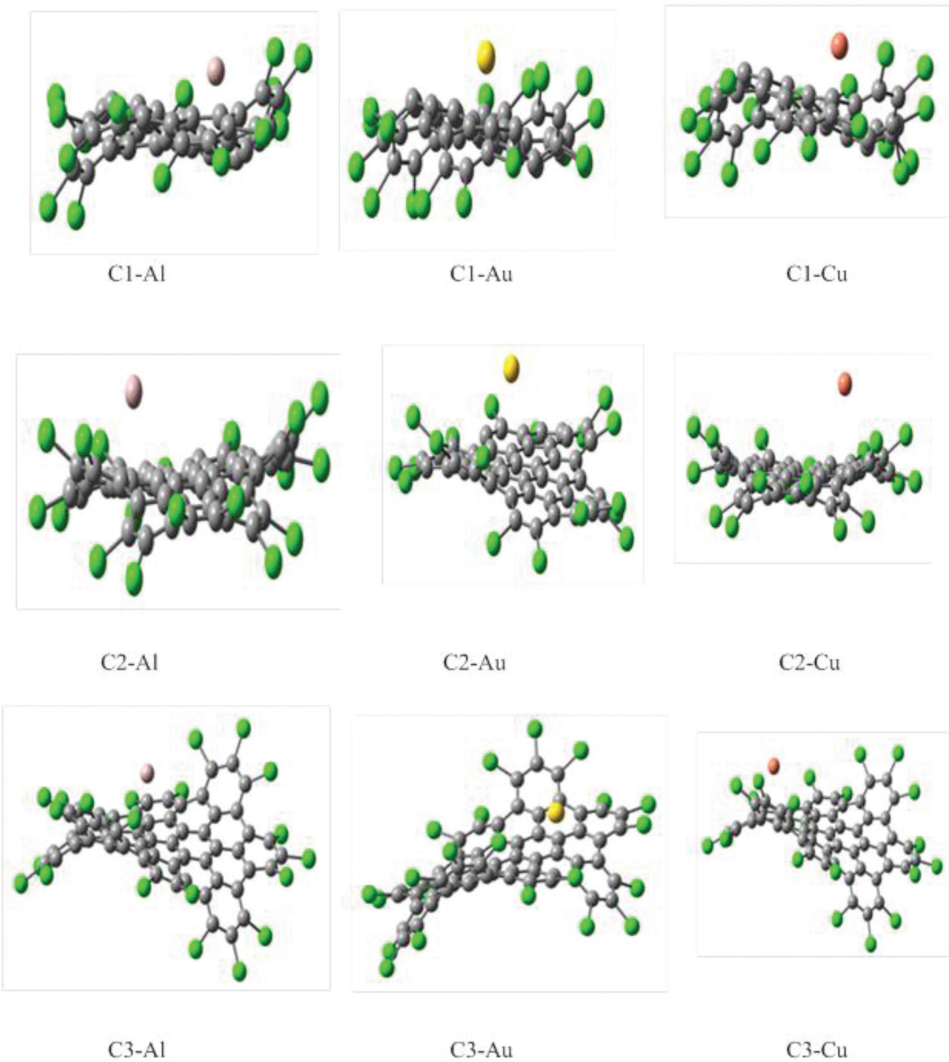

Figure 6. Potential adsorption sites and the most stable structures for Al, Au and Cu (dimers) on C1 ($C_{42}CI_{18}$), C2 ($C_{48}CI_{18}$) and C3 ($C_{60}CI_{22}$). Adapted from Ref. [117].

4. Conclusions

In this chapter, we have discussed the role of DNA and CNTs in the field of green electronics. Metal nanoparticles and its interaction to the nucleobases and pure carbon aggregates are also described in detail. Current and future investigations of green nanotechnology will provide a more complete knowledge regarding various factors that influence green synthesis of nanoparticles and the most sophisticated technology that can be used for characterization of the synthesized nanoparticles for its more efficient future applications in environmental, optoelectronic and biomedical field.

"United Nations World Commission on Environment and Development" has stated that the sustainable development is established when humanity ensures its present needs without compromising the ability of future generations to meet their own needs. So the time has come when we have to bear the responsibility for the shape and type of environment our future generations will live in; a healthy environment, a non-toxic world. Right now the world is not what we have set up for the mankind. Since electronics has now become an indispensable part of our life- for us and our future generations. The natural and nature inspired materials allow the "green" technologies to achieve the substantial goals in the electronics field: they embody low energy and have biodegradable and biocompatible materials as their backbone. Now the time has started to imagine and explore the new possibilities of "Green" organic electronics.

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