Chapter 2
Background of the Study

Abstract CNTs are allotropes of carbon in the nano form of a cylinder. In this chapter, the history of generating CNTs, the applications, and chemical properties of CNTs are reviewed. Also, the different generation of glucose biosensors as well as CNTs-based nanocomposite for electrochemical detection of glucose are highlighted. Generally, nanostructures have great potential to explore as simple and easy blood glucose checking medical tools. Electrochemical based oxidation of the immobilized enzymes (glucose oxidase) on CNTs has opened exciting opportunities for the determination of glucose. According to the previous publication related to the CNTs-based biosensor, CNTs display a perfect combination of electrical properties which enhance the sensitivity and selectivity of the fabricated amperometric glucose biosensors.

2.1 Structures of Carbon Nanotubes

The study reported by Saifuddin [1] highlighted three different possible types of CNTs namely; zigzag carbon nanotubes, chiral carbon nanotubes, and armchair carbon nanotubes. In fact, the method of rounded rolled axis of the graphene sheet's hexagonal network and the diameter of the closed end cylinder. The study presented by Lehman [2] also provides another classification of carbon nanotubes into SWCNTs and MWCNTs. Mostly, SWCNTs are more prone to defects during functionalization compared with MWCNTs. In terms of characterization and evaluation, this is easier with SWCNTs than MWCNTs [3]. SWCNTs are also simply pliable in contrast to MWCNTs which are not easily pliable. SWCNTs show resistivity in the range of 10^{-4}–10^{-3} Ω while the resistivity range of MWCNTs is from 1.8×10^{-5} to 6.1×10^{-5} Ω owing to the absence of defects in the structure. The purity of MWCNTs is higher since many studies highlighted that the purity of a typical MWCNTs depends on its synthesizing method [4].

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2.2 Generation of Carbon Nanotubes

Different morphology of CNTs has been reported based on powder form and Thin film due to their proposed application. Since the discovery of CNTs by Iijima [5], much attention has been attracted to this novel nanostructured material. Different synthesizing methods have been investigated to grow powder/thin film CNTs, including arc discharge [6], laser vaporization [7], and CVD [8]. In this section, some of the popular growing methods of CNTs are discussed in detail [9].

2.2.1 The Arc Discharge Method

Generally, the arc discharge method is one of the most practical synthesizing methods of CNTs for scientific purposes. Arc discharge yields highly graphitized CNTs due to a high temperature synthesizing process [10]. With this method, a direct current (DC) voltage is provided across the two electrodes (graphite) which are immersed in inert gas. When a pure graphite rod is utilized, a quantity of soot fullerenes are deposited inside a chamber, and MWCNTs are localized on the cathode. Ando [11] demonstrated that the growing CNTs followed by the DC arc discharge between a silicon anode and a graphite cathode. Anodic silicon is attached to a graphite rod. Then, the block of the silicon was removed and the DC arc is continuously discharged between the electrodes (approximately 1–2 mm apart). Ando suggested that there is a large amount of a soot carbon was discharged with fullerene. In addition to this result, MWCNTs were investigated as an additional carbon remained at the negative graphite electrode.

2.2.2 The Laser Vaporization Method

This method is also used to grow CNTs especially for synthesizing SWCNTs since the carbon target is ablated by a very high energy extracted from the laser beam. The collected results highlighted by Lebedkin [12] show that SWCNTs were grown by different diameters using laser vaporization of carbon rods which were doped with Co, Ni, and FeS (in an atmosphere of $Ar:H_2$). The results illustrate that the temperature and the gas pressure in the laser vaporization directly affects the diameter distribution of the grown CNTs. The obtained results were also represented that by the laser vaporization of a FeS: Ni: Co: C target in the $Ar:H_2$, with large diameters of tubes broadly distributed from ~ 2 to ~ 5.6 nm were obtained.

2.2.3 *The Chemical Vapor Deposition Method*

Nowadays, CVD is the most popular method for synthesizing CNTs, due to its ability to produce different CNTs by varying length of nanotube [13], uniformity of orientation [14], purity [15], and low cost [16]. The main part of this synthesis is to exhaust ahydrocarbon vapor for a period of 15–60 min via a tubular reactor. Inside this reactor [17] is a metal catalyst which can accommodate temperature in the range of 180–400 °C on which SWCNTs or MWCNTs can be grown. These will be extracted after the cooling system has reduced the catalyst to the room temperature. Where liquid hydrocarbons are used as a carbon source, the catalyst will be heated while an inert carrier gas carries the hydrocarbons. Hence, the hydrocarbon vapors are transported to the reaction zone of the CVD. Solid hydrocarbon needs time to become usable as the CNTs precursor in a low temperature zone within the reaction tube. The implication drawn from this synthesis is that hydrocarbon vapor initially decomposes into two elements namely carbon and hydrogen [15]. This decomposition happens when the hydrocarbon vapor touches the hot catalyst nanoparticles. Thus the hydrogen leaves and the carbon dissolves into the particular metal [18]. Figure 2.1 represents the schematic of the horizontal CVD setup described above.

2.2.3.1 Main Parameters on Carbon Nanotubes Produced by Chemical Vapor Deposition Method

It has been found the CNTs morphology grown by CVD depends on the most effective parameters in the synthesizing process. A perfect control of the amount of the flow rate of the carrier inert gas, amount of the catalyst and precursor, the reaction time and its duration, applied temperature, and annealing system allow CVD to produce the interested structure of CNTs. A number of research groups reported that selective and controlled growth of MWCNTs by varying the effective parameters in fabrication process is necessary for their proposed applications [19].

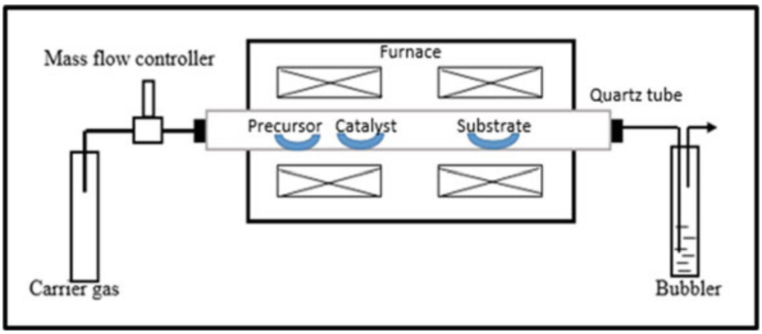

Fig. 2.1 Schematic setup of the horizontal CVD

However, to maintain consistency with the objective of this book, we will consider some key parameters in the following.

2.2.3.2 Effects of Temperature on Carbon Nanotubes Growth

The main aspect of the relationship between temperature and the growth of CNTs is the increase of thermal resistances when the CNTs arrangement growth temperature is decreased [17]. In the study by Cola [20], thermal resistances of silicon-CNT-silver interfaces grown through CVD can accommodate temperatures ranging from 506 to 806 °C where the temperature was measured via the photoacoustic technique. The results of this study showed that CNTs arrangements which offer fine heat interface conductivity could be grown when temperatures are reduced. When the temperatures are reduced, there is a possibility of integration with some sensitive substrates. The measurement of thermal resistance showed that it increased when the CNTs array growth temperature was reduced. In other research on the effect of the temperature in the CVD method, Amama [21] found that the CNTs growth rate (length of CNTs/duration of the process) goes up by four times by increasing the growth temperature. In a meantime, the average diameter of the grown CNTs was doubled with an increase in temperature [22].

2.2.3.3 Effects of Carrier Gas Flow Rate on Carbon Nanotubes Growth

Flow rate of the carrier gas also has an impact on the growth of CNTs and this is shown in the study by Reynolds [23]. The authors conducted research on the synthesis of CNTs as well as their characteristics when used on silicon substrates. The synthesis of CNTs in this study involves the CVD method at a temperature of 900 °C with the aid of methane and hydrogen flow—different flow rates of hydrogen gas were used to optimize the CNTs' yields. Their results illustrated that hydrogen gas at a flow rate of 100 sccm yields a very low quantity of CNTs. However, hydrogen at a flow rate of 200 sccm yields a slightly higher quantity of CNTs but still a limited amount. However, a hydrogen flow rate of 300 sccm yields plentiful CNTs with a smaller outside diameter and shorter branches. From these results it is concluded that the quantity of CNTs increases when the flow rate of hydrogen is increased from 100 to 300 sccm and the density of CNTs is at its peak when the flow rate of hydrogen is in the range of 300–400 sccm.

2.2.3.4 Effects of the Annealing and Vaporization Time on Carbon Nanotubes Growth

The diffusion of the hydrocarbons and the duration of the annealing step have a significant effect on the length and the diameter of grown CNTs. In fact, the aspect ratio of synthesized SWCNTs or MWCNTs increases by increasing the durations of CVD, especially at longer vaporization and annealing times. This alteration in the diameter or the length of the synthesized CNTs might be due to sintering and the agglomeration of the feedstock. The results reported by Kim [24] show that on top of the reaction temperatures in the CVD method, the aspect ratio of CNTs is directly affected by the application of multiple reaction times. In cases of longer reaction times, both the diameter and length of the grown CNTs will also increase. The results also confirmed that the reaction time adapts to CNTs growth. In other words, the diameter of the grown CNTs is determined by the duration time of tubes in the hottest zone of the CVD reactor. Choi [25] reported that the diameter of a CNT was a function of its growth rate (length of the CNTs/reaction time). Accordingly, the length of CNTs increases with reaction times, clearly showing that the development of the structure of CNTs continues up to the point that the hydrocarbons are depleted.

Controlling annealing time is one of the main factors affecting the quality and yield of CNTs, as highlighted in Park [26]. Based on the obtained results from low deposition temperatures, in samples annealed for 20 min some remaining carbonaceous particles were removed from the CNTs' surface, and the total weight was reduced by $\sim 70\%$. By increasing the annealing time to 40 min, a major amount of the exciting carbonaceous particles was removed and the weight was reduced to $\sim 35\%$. The author also pointed out that although the relative population of nanotubes to carbonaceous particles should increase more with increasing annealing time, extended annealing times will result in overburning of nanotubes.

2.2.3.5 Effects of Catalyst on Carbon Nanotubes Growth: Ferrocene

Compared to the numerous methods developed to synthesize CNTs, catalytic CVD has the tendency to produce nanotubes with fewer impurities and nanotubes produced via this method appear to be amenable to large scale low cost processing [27]. According to Liao [28], there are few theories developed about the way CNTs grow from catalyst particles when CVD is conducted. One of the predicted theories described in the above study holds that CVD using catalysts involves the process of decomposition of gases containing carbon such as hydrocarbons or carbon monoxide. This decomposition occurs on one of the sides of a very small catalyst particle followed by the diffusion of carbon atoms via the particle's surface. Eventually, precipitation of CNTs occurs in the second furnace. The research based on this theory found that the size of metal-based catalysts will impact the growth rates of CNTs. For instance, sometimes the carbons have different morphologies but the catalyst particles can be alike in sizes. But in their experiment the researchers found that a catalyst with much larger particles was able to catalyze some long CNTs formation. The most common catalysts used for the CVD method are iron, cobalt, and nickel due to the perfect solubility and excellent diffusion rate of carbon in these metal particles [29, 30]. Some hypotheses state that carbon morphology changes when the temperature used for synthesis is varied [31]. Researchers perceive that a potential factor in the carbon morphology is the constitution of the catalyst. Kumar [32] reported that iron has a high catalytic effect in hydrocarbon decomposition during the synthesizing process of CNTs. However, using cobalt as a catalyst resulted in well-graphitized CNTs but lower CNTs yield than using iron. On the other hand, the appropriate concentration of catalyst also plays an important role in the desired growth rate of CNTs.

Carbon nanotube mechanistic models described by Neil [33] clearly show both the mechanisms used to illustrate the catalytic growth of CNTs on the substrate. According to the results, in the tip-growth mechanism (Fig. 2.2a), the catalyst is located at the top of a growing nanotube. But in the base-growth mechanism (Fig. 2.1b), the catalyst is located at the bottom of the nanotube. In both mechanisms, the carbon reagent decomposes on the catalyst under the reaction process. The results also demonstrate that the interaction between catalyst and CNTs in the base-growth mechanism is stronger than in the tip-growth mechanism.

According to Asli [34], if a lower quantity of ferrocene is applied, a lower yield of CNTs will be obtained. There will also be lesser bend and multidirected growth. Another observation is that nearby nanotubes do not support each other resulting in random orientation of the CNTs. The study observed that with less than 0.4 g ferrocene, there is no indication of aligned CNTs production. However, when the quantity of ferrocene was multiplied, the CNTs production improved stage by stage. On the CNTs produced by higher amounts of ferrocene, carbonaceous crusts were visible covering the upper part of the CNTs. The authors attribute the presence of carbonaceous crusts to the greater quantity of ferrocene relative to the quantity of carbon supplied and the lack of uniformity in the evaporation of the ferrocene and

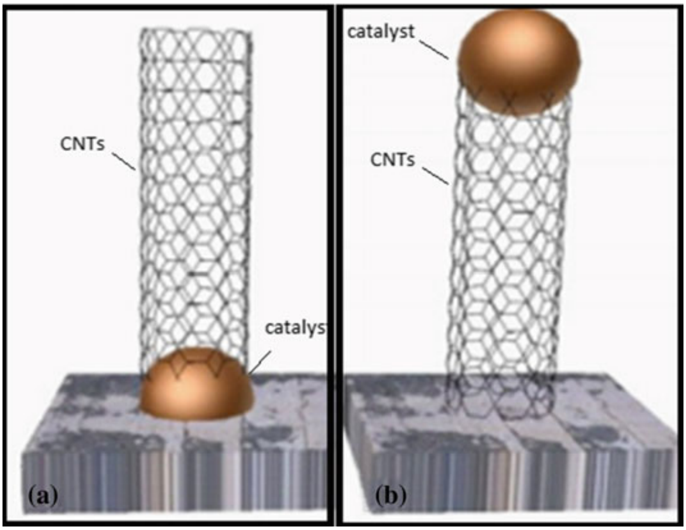

Fig. 2.2 Carbon nanotube mechanistic models: **a** Base-growth mechanism; **b** Tip-growth mechanism [33]

camphor oil during the synthesis. When they experimented using 0.5 g of ferrocene and 5 g of camphor oil, CNTs appeared to have a dense configuration with uniformly shaped vertical columns. This experiment demonstrated the formation of CNTs with the length of 110–113 μm. It also showed a growth process of CNTs in vertical columns providing support for each in order to continue growing in the same direction, at 90° to the template surface. Thus the implication is that lower quantities of catalyst can produce CNTs lower in density. The researchers summarized their findings by stating that CNTs growth can be observed even with a quantity of ferrocene as low as 0.3 g. However, to produce VACNTs, the minimum amount of ferrocene required is 0.4 g [32].

2.3 Fabrication of Glucose Biosensor: The First, Second, and Third Generation

Diagnosis and management of *diabetes mellitus* needs unique investigation and blood monitoring for glucose. Many patients are suffering from sugar related diseases, such as diabetes, which requires them to check their blood glucose levels every day. This makes glucose one of the most common blood analytes for testing. Hence, glucose biosensors account for about 85% (enzymatic biosensors) of the whole biosensors market. This huge demand and potential market size leads the development of new biosensing concepts [35]. Much of the interesting research and novel strategies for glucose detection by biosensors has led scientists to improve and develop cheap and easy-to-use glucose biosensors. Enzyme electrodes, especially electrodes based on GOx, have played a leading role in the move to simpler and easy-to-use methods for monitoring blood sugar [36].

Lower dynamic ranges as well as susceptibility to variations in oxygen concentration in the solution are some of their main disadvantages of the first generation of glucose biosensors. These rely on detection of hydrogen peroxide and the use of natural oxygen cosubstrate [37]. Reactions based on biocatalytic processes involve reduction of the flavin group (FAD) in the enzyme by reaction with glucose to produce the reductive form of the enzyme ($FADH_2$) [38].

The second generation of glucose biosensors rely on detection of the glucose level in the buffer solutions via enzymes which can catalyze the glucose. However, due to the thick layer (protein), enzymes do not transfer the electrons directly to counter electrodes. Different approaches have been investigated for monitoring the existing electrical contact of the redox center of GOx and the last surface of the electrode [39]. Measuring the catalytic current arising from the reoxidation of the mediator is the principal mechanism of the second generation of glucose biosensors [40]. In this regards, several types of electron mediators could be used as electron shuttle in the system [41]. Thanks to the presence of the mediator, greater linear detection range is one of the advantages of the second generation of glucose biosensors. They are also less susceptible to electroactive interference because of their ability to operate at lower potentials [42].

In the third and latest generation of biosensors the exited electron from the detection process is transferred directly to the electrode from the analyte [43]. Omitting mediators from the system is one of the advantages of these biosensors, making them an excellent selective with low operating potential biosensor [44].

2.3.1 Carbon Nanotube-Based Biosensors

CNTs are one of the best materials for use in different sensing tasks due to their electrical properties and functional possibilities. Beside the aspect ratio of CNTs, chemical functionalized can also be a useful to immerse any chemical species to surface of the CNTs which is improving the solubility and biocompatibility of the CNTs. This can allow the realization of composite electrodes comprising CNTs well dispersed in an desired matrix of polymers [45, 46]

2.3.2 Functionalization of Carbon Nanotubes

The functionalization methods of CNTs include non-covalent and covalent techniques which can improve the interactions of nanotubes with solvents [47]. Covalent functionalization is defined as a linkage of functional entities by changing hybridization from sp^2 to sp^3 which can take place at the terminus of the tubes as well as the on the sidewalls of them [48]. Recent published reports show that many methods can improve the possible application of CNTs using functionalization methods which include: oxidative purification based on carboxylation [49], defect functionalization [50], halogenation [51], hydrogenation [52], addition of radicals [53], addition of nucleophilic carbenes, sidewall functionalization through electrophilic addition [54], cycloadditions [55], and aryl diazonium chemistry [56]. Non-covalent functionalization consists of complexation of supramolecules through various attractive or repulsive adsorption forces, such as π–π stacking interactions and van der Waals [57]. Results investigated by Gorton [58] and Challa [59] clearly demonstrated that functionalized CNTs enhance the electrochemical reactivity of biomolecules and can improve electron-transfers in proteins with embedded deep redox centers. Therefore, a variety of studies were focused on using the remarkable advantages of functionalized CNTs in electrochemical sensing applications such as clinical diagnostics or environmental monitoring. Alcohol, amino acids, cholesterol, lactate, pyruvate, glucose, glutamate, and hydroxybutyrate are some of the important clinical analytes to generate electrochemically detectable products. The focus of this book is the glucose monitoring process which consists of common chemical functionalized methods of CNTs for immobilization of GOx to fabricate electrochemical glucose biosensor based on CNTs.

2.3.3 Functionalized Carbon Nanotubes to Enhance Electron Transfer Mechanism in Glucose Biosensor

Monitoring of blood glucose is conducted routinely in medical practice [60]. In almost all of the developed countries about 5% of population have a diabetic condition. Thus the detection of glucose in patients' body fluids had been a major activity for doctors [61]. Glucose sensors usually make use of GOx in the detection of glucose. GOx is an enzyme where it functions to catalyze the oxidation process of β-D-glucose to D-glucono-1, 5-lactone and oxygen (O_2) used as the electron acceptor. From this process, a new combination hydrogen peroxide (H_2O_2) is produced. Hydrogen peroxide is electrochemically identified on a suitable electrode. In these processes, GOx displays great specification for β-D-glucose compared to 2-deoxy-D-glucose, D-mannose, and D-fructose which are oxidized at a lesser turnover [62]. The conductivity of CNTs as well as their small size of them makes them suitable as nanoelectrodes in the glucose detection process. Many researchers have focused on promoting the electron transfer reactivity of CNTs in biosensors. Table 2.1 shows some of the recent research reports on glucose biosensors based on functionalized CNTs enhanced by the electron transfer method.

Fabrication of a biosensor using Janegitz method can be easily conducted with precision and it has an accurate response for glucose determination as shown in Fig. 2.3. According to the obtained results it can be concluded that determination of glucose using a glucose oxides-carbon nanotube-dihexadecyl hydrogen phosphate-glassy carbon electrode (GOx-CNTs-DHP-GCE) as a detecting electrode of the biosensor had better analytical characteristics than those described by Li [71] and Wu [72].

Pandey [66] had reported similar findings on VACNTs which show better stability of the synthesized glucose biosensor. According to their results, CNTs were electrochemically functionalized with carboxylic (COOH) group acids. The modified electrodes were then dried in the air. Immobilization of GOx, was performed by immersing CNTs in a solution which contained coupling agents EDC (1-ethyl-3-3-dimethylaminopropyl carbodiimide) and Sulfo-NHS (N-hydroxy-sulfo-succinimide) for 3 h at 4 °C. A cyclic voltammogram of GOx functionalized CNTs clearly indicates a pair of symmetrical redox peaks corresponding to the electron transfer reaction of GOx as shown in Fig. 2.4. The results indicate the limit of detection, which was estimated around 14.4 μM with the sensitivity of 410 nA mM^{-1} cm^{-2} by considering the sensor active area of 0.385 cm^2.

Raicopol [64] published a report using another new type of glucose biosensors by using polypyrrole (PPy). In his report, CNTs were uniformly dispersed in deionized water, and then the suspension solution was transferred to a flask containing sulfanilic acid and tert-butyl nitrite. After mixing, an appropriate amount of acetone was added and then CNTs were filtered. In the last step, the CNTs were treated with a diluted NaOH solution and again filtered, washed with water followed by acetone, and dried. The selected amount of GOx was then added to the CNTs solution (functionalized nanotubes were dispersed in bidistilled water), and then pyrrole was added to the GOx-CNTs-$PhSO_3$ mixture, followed by

Table 2.1 Glucose biosensor based on functionalized CNTs improved by immobilizing CNTs

Nanoelectrode nanoensembles	Functionalizing method of CNTs	Immobilizing method	Limit of detection	Sensitivity	Stability (approx)
GOx-CNTs-DHP [63]	Pretreatment sulfuric and nitric acids	Carboxyl coupling reaction using EDC and N-hydroxysuccinimidyl (NHS)	9 μM	Not mentioned	One month
PPY/GOx/SWCNTs-$PhSO_3^-$/PB [64]	Flask fitted with a magnetic stirrer using sulfanilic acid and tert-butyl nitrite	Electropolymerization of GOx in dispersed CNTs	0.01 mM	6 μA mM^{-1} cm^{-2}	Not mentioned
PyBA-CNTs-GOx [65]	Modified using 4-(pyrrol-1-yl) benzoic acid (PyBA)	Embedding by composite film	10.2 mM	Not mentioned	Less than a month
VA-MWCNTs-GOx [66]	Sonication by HNO_3 and H_2SO_4	Immersing method using EDC and Sulfo-NHS	14.4 μM	410 nA mM^{-1} cm^{-2}	Six months
TP/MWCNT/GOx [67]	Sonicating in a mixture of concentrated sulfuric and nitric acids	Electrostatically deposited by stirring MWCNTs and GOx in PBS	Not mentioned	Not mentioned	Not mentioned
GOx/MWCNTs [68]	Carbodiimide coupling reactions	Covalent method	12 mM	20.6 μA mM^{-1} cm^{-2}	One week
CP- GOx-CNT_{EF} [69]	Electrooxidizing method using HNO_3	Physisorption of GOx based on soaking method	10 mM	Not mentioned	Not mentioned
PDDA/GOx/PDDA/CNTs [70]	Sonication by HNO_3 and H_2SO_4	Adsorption by dipping technique	7 μM	15 μA mM^{-1} m^{-2}	Four weeks

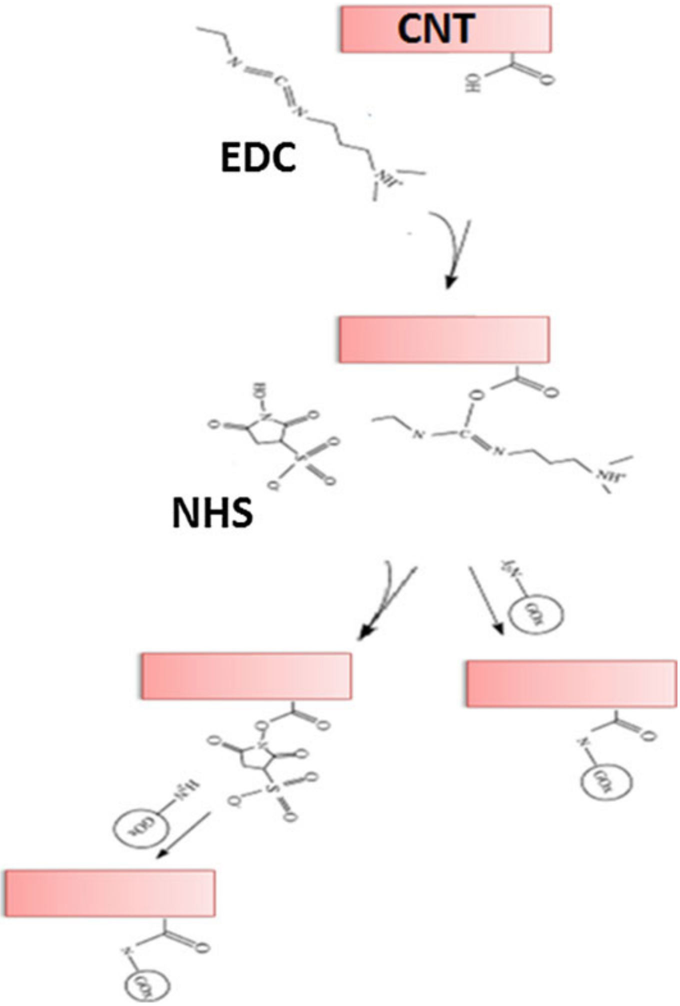

Fig. 2.3 Scheme of the reaction of 1-ethyl-3-3(dimethylaminopropyl) carbodiimide (EDC) and N-hydroxysuccinimidyl (NHS) with CNTs and GOx [73]

electropolymerization in pH 7 and the sample was subjected to overoxidation by cycling the potential in PBS solution as indicated by Fig. 2.5.

Self-assembling of GOx-based biosensors on the surface of CNTs was reported by Liu [70]. In this “flow injection method”, the CNTs were firstly functionalized by ultrasonication with H_2SO_4 and HNO_3 and then dried in the vacuum. Then, the functionalized CNTs were immediately dispersed in N,N-dimethylformamide following by dropping on the GCE. The results show that fast current responses and well-defined detection were obtained for different glucose concentrations. The low noise level and high sensitivity allow the detection of micromolar concentrations in

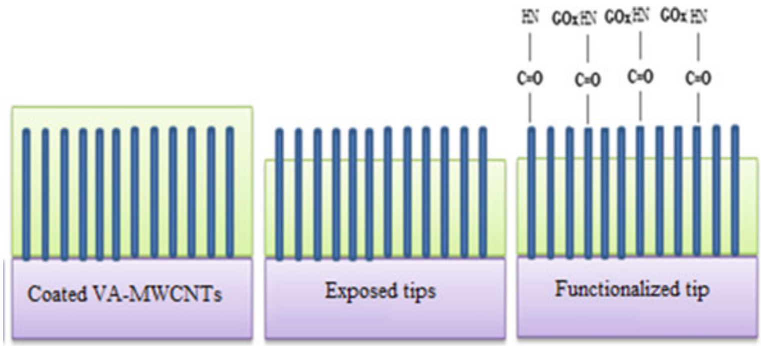

Fig. 2.4 Schematic diagram of the fabrication steps of CNTs nanoelectrode nanoensembles (CNNEs). VA-MWCNTs dipped and coated with poly methyl methacrylate (PMMA). The polished PMMA coated VA-MWCNTs with exposed tips and CNNEs functionalized with GOx [66]

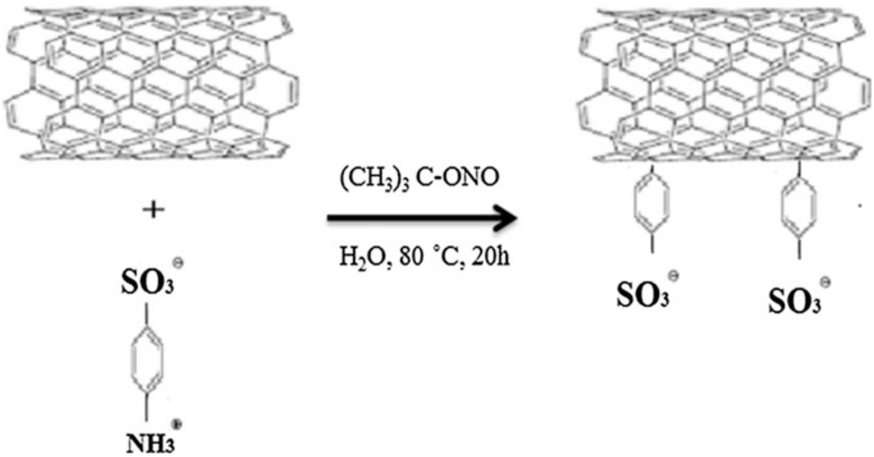

Fig. 2.5 Functionalized single-walled carbon nanotubes (SWCNTs) [64]

the range 15–120 µM. Figure 2.6 shows a schematic of wall-jet flow cell and upper piece of the wall-jet flow cell.

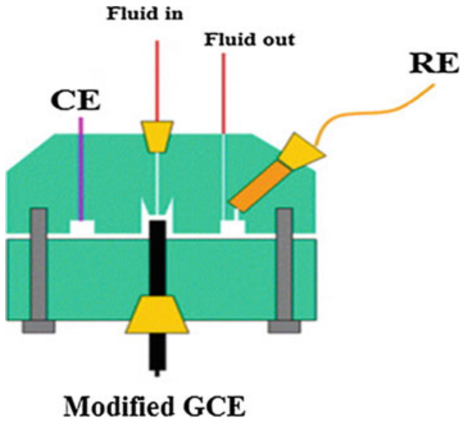

Fig. 2.6 Scheme of homemade wall-jet flow cell [70]

2.4 Carbon Nanotube-Based Composites in Glucose Biosensors

As shown by the previous results in Table 2.1, the incorporation of enzymes with SWCNTs and MWCNTs allows the fabrication of selective and sensitive enzyme-based CNTs biosensors. In this regard, CNTs nanocomposite and some other possible materials such as nanoparticles can be used to enhance selectivity and sensitivity of the fabricated biosensors as shown in Fig. 2.7 [74].

The use of CNTs-nanocomposites is another fabrication method for glucose biosensors that follows the entrapment technique which can be defined by its stability, sensitivity, and detection limit [75]. Table 2.2 summarizes some of the recent fabrication methods of nanocomposite-based CNTs for glucose biosensors.

From the data in Table 2.2 it can be concluded that the CNTs nanocomposite-based biosensors that incorporated nanoparticles are responsible for producing higher sensitivity than the conventional glucose biosensors. Attribution of this can be made to the improved catalyst functions, fine biocompatibility, and the very large surface area produced.

2.5 Functionalization of Carbon Nanotubes for Other Applications

Functionalized CNTs is one of the effective methods of enhancing the solubility of the CNTs which allows researchers to modify the CNTs for their potential applications. The functionalization methods include covalent and non-covalent functionalization which can improve the interactions of nanotubes with polymers or solvents [47]. Covalent functionalization is defined as a linkage of functional

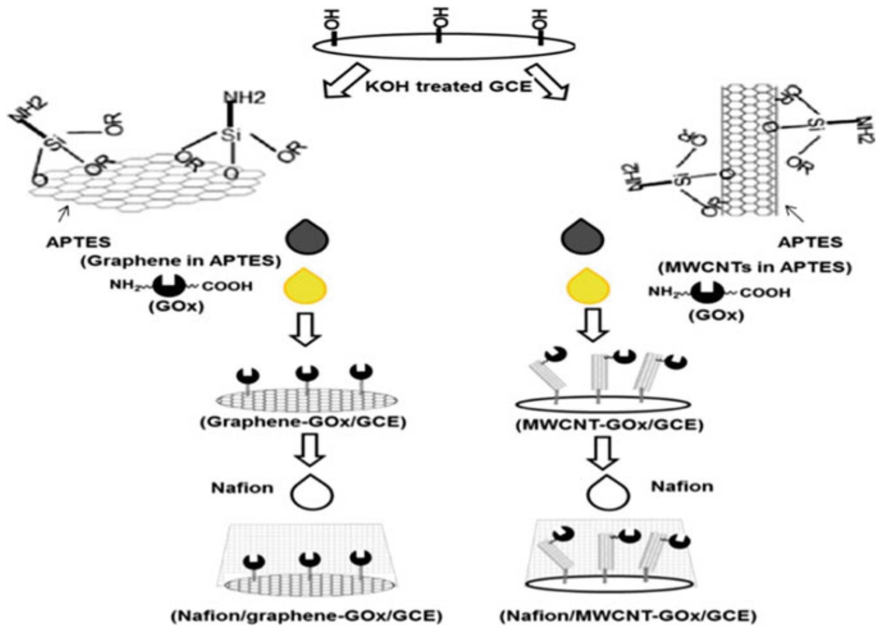

Fig. 2.7 Schematic configuration of graphene versus MWCNTs for electrochemical glucose biosensing [76]

entities by changing hybridization from sp^2 to sp^3 which can be performed at the termini of the tubes as well as on the sidewalls of it [48].

2.5.1 Thermal Applications of Carbon Nanotubes

The history of CNTs and their various applications discussed above lead us to questions about the suitability of using CNTs in those applications. Studies of the physical and chemical properties of CNTs by many researchers are essential in identifying application for which CNTs will be appropriate. Since the discovery of CNTs, this material been applied in various fields for various purposes. One such applications is described by Fabris [89]. The researchers concluded that improvements in thermal interface materials (TIMs) can enhance heat transfer in electronic packages and reduce high temperatures. In their experiment, two types of TIMs were tested based on the addition of CNTs. The first one was mixed with a commercial TIM product and the other one made of only CNTs and silicon oil. After that the materials were tested using an in-house apparatus that allowed for the simultaneous measurement of temperature, pressure, heat flux, and TIM thickness. The results show that CNT-thermal grease mixtures are more compliant, with a small increase in bulk thermal conductivity over the range of tested pressures [90].

Table 2.2 The key parameters in CNTs-based nanocomposites for glucose sensors

Fabrication method	Sensors	Sensitivity $(mmol/L)^{-1}$	Linear range of detection (mmol/L)	Response time (s)	Stability
Nanoparticle decoration, Enzyme adsorption	Nafion/GOx/PtNP/CNT/Graphite [77]	14 µA	0.1–13.5	<5	73% after 22 days
Nanoparticle decoration	CuNP-SWCNT-Nafion/GCE [78]	256 µA	0–0.5	10	Not mentioned
Electro polymerization	PPy-GOx-MWCNT array/Gold [79]	350 nA	2.5–20	Not mentioned	70% after 3 days
Encapsulation	GOx-Nafion-MWCNT/GCE [80]	330 nA	0–0.002	<3	
Electro polymerization	POAP-GOx/FePc MWCNT [81]	735 nA	0.0005–4	<8	120 days
Nanoparticle decoration, Enzyme adsorption	GOx/PtNP-SWCNT-Nafion/GCE [82]	2.11 µA	0.0005–5	3	
Electro polymerization	PPy-GOx-MWCNT/GCE [83]	2.33 nA	0.2–50	15	Not mentioned
Encapsulation, sol-gel process	GOx-SGC/MWCNT/bppg [84]	196 nA	0.2–20	<5	3 weeks
Adsorption	GOx/Au/Pt/CNTs [85]	21 nA	0.4–11.89	5–7	25 days
Electro deposition	Nafion/GOx/Pt/MWNTs [86]	58.9 µA	1–23	5	50 days
Electro deposition	Pd-GOx-Nafion CNTs [87]	Not mentioned	12	3	14 days
Electro deposition, sol-gel	CNT-PtNP-CS-MTOS [88]	4.94 µA	1.2–6	5	30 days

2.5.2 Medical Applications of Carbon Nanotubes

Because of their properties CNTs have found many biomedical applications. Wu [91] found that CNTs have created much interdisciplinary interest due to their unusual structural, mechanical, and electronic properties. The authors of this research first reviewed the covalent surface chemistry for CNT functionalization and then studied the surface chemistry of CNTs for biomedical applications, especially in relation to controlling biomedical functions. The outcome showed that CNTs had exactly the needed chemistry for use in biomedical applications. CNTs are predominant in today's medical research domain and are being thoroughly researched in the areas of efficient drug delivery, biosensing approaches for treatment of disease, and health screening. Growing knowledge of CNTs has revealed the possibility of modifying drug delivery as well as biosensing approaches in various fields of medicine.

The application of CNTs in the field of drug delivery and biosensing has revolutionary potential in medicine. Functionalization of SWCNTs has improve the solubility rate allowing for effective tumor targeting or efficiently drug delivery. It also avoids the threat of SWCNTs displaying cytotoxic behavior and modifying the role of immune cells.

Cancer, a type of disease in which cells in the body or at a certain organ develop and divide in an abnormal way, is one of the main diseases being focused on with regards to the way it reacts to CNT drug delivery. Present cancer therapy mainly includes surgery, radiation therapy, and chemotherapy which normally cause pain, destroy healthy cells, and produce opposing side effects. As drug delivery transporters, CNTs have displayed the capability of targeting certain cancer cells with a lower quantity of drugs than usually applied—which is equally efficient in destroying the cancer cells, does not harm healthy cells, and drastically reduces side effects. Zhang [92] provides vital information and knowledge about the usage of CNTs in cancer treatment.

The study revealed that CNTs have fulfilled three main requirements making them ideal carriers for targeted drug delivery: they themselves have target effects; they have sufficiently strong adsorptive effects for anticancer drugs to ensure they can transport the drugs to the effect-relevant sites; and they can release the drugs from them in the effect-relevant sites [93].

Among the above approaches, Zhang [92] found that functionalization has been the most effective. In addition, functionalization has been shown as capable of decreasing cytotoxicity, improving biocompatibility, and giving opportunity to appendage molecules of drugs, proteins, or genes for the construction of delivery systems.

According to Abdelbary, CNTs are applied in cancer therapy and drug delivery [94]. CNTs have been studied as possible nanocarriers for the delivery of drugs, genes, and proteins. Most of the study on CNTs has centered on their potential as deliverers of anticancer agents. This function is possible due to the needle-like shapes of CNTs which is unique, allowing CNTs to absorb or bond covalently to a huge range of therapeutic materials and deliver them internally into the aimed cell. This study further discussed the application of CNTs as carriers of anticancer molecules because it is widely known that cancer cells overly express folic acid receptors and some research groups have made nanocarriers with engineered surfaces to which folic acid derivatives can be attached. The research also provided the information that non-spherical nanocarriers have been discovered to stay in the lymph nodes for longer periods of time compared to spherical nanocarriers. CNTs are examples of non-spherical nanocarriers and as such may have potential use for destroying lymph node cancers [95].

The study summarized that the capability of macromolecules to move against biological obstacles and be conveyed into a target cell is especially limited, because of their hydrophilicity and large molecular size. The researchers in this study also added that gene therapy targets to utilize genetic substances to medicate diseased cells by mending the reason of the disease. Due to the reason genetic substances are unable to bypass the biological membrane the application of viral or non-viral vectors to bring the gene and suppress it inside the cell is highly needed. Non-viral vectors are inefficient compared to viral vectors and have a shorter lifespan; nevertheless, these non-viral vectors are safe.

According to the researchers of this paper, previous researchers have established original functionalized SWCNT-DNA multiplexes and found that more DNA manifestation when compared with naked DNA [96]. Commonly, functionalized SWCNTs have been recommended by this study as appropriate non-viral carriers of macromolecules and internalization of those macromolecules within living cells by CNTs has been found to take place via energy-based endocytosis. The finding showed that confocal microscopy and flow cytometry have displayed much bigger fluorescent movement of protein and DNA when conjugated to SWCNTs compared to the naked macromolecules which shows that CNTs are prominent vectors for gene and protein. According to the authors, other researchers have presented a method for gene distribution called "carbon nanotube spearing" [97]. Carbon nanotube spearing consists of a process where plasmid DNA with a fluorescent protein was immobilized onto nickel-embedded CNTs. This was "speared" into Bal 17 B lymphoma cells with the aid of a magnetic field, which makes for a high rate of transfection in the target cells.

2.5.3 Antimicrobial Activity of Carbon Nanotubes

Other than cancer, other research reported therapeutic applications of CNTs where there is a possibility for surface-engineered CNTs to be able to capture pathogenic bacteria in a liquid medium [98]. Therefore, CNTs themselves may have antimicrobial process possibilities since microorganisms may be adsorbed into the engineered surfaces of CNTs. For examples, utilizing *Escherichia coli* as a sample microorganism, it has been discovered that the electronic attributes of SWCNTs might control their antibacterial process. The antibacterial property was a result of

CNT-induced oxidation of the intracellular antioxidant glutathione [99] causing increased oxidative stress on the bacterial cells and eventual death of the bacteria [100].

2.5.4 Drug Delivery Applications of Carbon Nanotubes

The researchers added that functionalized CNTs have the perfect ability to act as carriers for antimicrobial biocomponents such as antifungal amphotericin B [101]. CNTs are able to be covalently attached to amphotericin B in order to bring it into mammalian cells [102]. The study reported the reduction of antifungal toxicity compared to the toxicity of the free drug: 40% of the cells were destroyed by the CNTs-free formulation compared to no cell death by the CNTs formulation [103]. It was also discovered that the antifungal activity was increased using the CNTs [104].

2.5.5 Electro-optical Properties of Carbon Nanotubes

Mansano [105] studied the electro-optical properties of CNTs. According to the published report, this system is based on a new concept of plasma generation: “a planar coil is coupled to an RF system for plasma generation.” This was used together with an electrostatic shield for plasma densification, thereby obtaining high-density plasmas. The CNTs were deposited using pure methane plasmas. The researchers used three methods for the surface modification of the sample.

According to their study, the electro-optical effects depend strongly on the preparation of the substrate as well as the deposition parameters of the CNTs. Their CNTs were highly aligned showing singular properties that could be a platform for many exciting applications.

2.5.6 Mechanical Properties of Carbon Nanotubes

Treacy [106] indicates that CNTs are predicted have appealing mechanical properties especially the high stiffness and axial strength. These two properties are due to their seamless cylindrical graphitic structure. So far it has not been possible to directly measure the mechanical properties of CNTs because of their tiny dimensions. In this study, the researchers approximated the Young’s modulus of isolated nanotubes by measuring (in a transmission electron microscope) the amplitude of their intrinsic thermal vibrations. They found that CNTs have remarkably high Young’s moduli, in the TPa range. Their high stiffness, together with their low density, shows that CNTs may be useful as nanoscale fibers in strong, lightweight, composite substances.

2.5.7 Gas Sensors Based on Carbon Nanotubes

Wang [107] reported that CNTs are from the group of fullerene structures. The results defined SWCNTs as a one atom thick layer of graphite rolled up into a seamless cylinder with a diameter of several nanometers, and length on the order of 1–100 μm. The study also reported that, thermally, CNTs have high stability in both vacuum and air. The authors' description of electrical properties shows that CNTs can be either metallic (conductors) or semiconducting, depending upon the tube diameter and the chirality—which is the direction in which the graphite sheet is rolled to form the tube [108]. The chirality is usually represented by an integer pair (n, m). Nanotubes with $n - m = 3j$ (j being a nonzero integer) are metallic (conductors) while all the others are semiconducting [109]. The dielectric property of CNTs is highly anisotropic due to their nearly one-dimensional structures, which may enable nanotubes to carry high currents with negligible heating effect [110].

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