



# Manufacturing of electrochemical sensors via carbon nanomaterials novel applications: a systematic review

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Received: 24 May 2022 / Accepted: 9 September 2022 / Published online: 1 October 2022  
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**Abstract** Most significant optical, mechanical, thermal, electronic, and chemical properties of carbon nanomaterials assist to fabricate the best electrochemical sensors with enhanced performance. Carbon-based nanomaterials electrochemical sensors are utilized for detecting the different analytes or targets, and also it promotes high electron–transfer kinetics of proteins. The remarkable sensitivity of various carbon nanomaterials, namely

fullerene, graphene, carbon nanotubes, and rGO, is suitable for fabrication of electrochemical sensors. The electrochemical sensors are very simple, reliable, and cost-effective used for analyzing the amount of active electroanalytes effectively. In this organized review, we are mainly exploring verities of carbon nanomaterials and smart electronic material graphene for further utilization of electrochemical sensors and extensive characterization techniques broadening on industrial scopes.

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This article is part of the topical collection:  
“*Nanoarchitectonics for Functional Particles and Materials*”.

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Guest Editor: Katsuhiko Ariga.

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**Keywords** Carbon nanomaterials · Graphene ·  
Electrochemical · Sensors

## Introduction: an overview of carbon material research avenues

Metabolic illnesses have become more widespread as living conditions have improved, owing to changes in dietary and lifestyle behaviors that damage health. Currently, metabolic biomarkers are routinely utilized for assessing diagnosis, therapy, and prediction of metabolic illnesses. To detect the biomarkers of metabolic disorders, electrochemical sensor technology is implemented as it is economical simplicity, real-time analysis. In order to better the performance of electrochemical sensors, carbon nanomaterials were used as promising materials. Carbon nanotubes, carbon nitride, graphene, rGO, and carbon quantum dots are the most common carbon nanomaterials used in electrochemical sensors for identifying the biomarkers of

metabolic disorders. The development of electrochemical biosensors are focused and drawing considerable attention due to its more durability, enhanced electrical conductivity, and biocompatibility [1, 2]. The electrical technology combined with advancement of nanomaterials has been accelerated large development in electrochemical biosensing technology which provides quick and easier way to detect the symptoms of metabolic illness. Carbon nanomaterials have been considered promising because of its unique properties like electrical, optical, mechanical, chemical, and thermal properties. The inherent properties of 0-D, 1-D, 2-D, and 3-D carbon nanomaterials influenced the great development and implementation of the advanced technology for sensing applications which includes fullerenes, CNTs, graphene and its derivatives, 0-D carbon quantum dots, nanodiamonds, and carbon nanohorns. Electrochemical biosensors based on carbon nanomaterials have been examined in the different segments such as improvement of sensitivity, selectivity, and detection range of various chemical and biological substances. This work presents an assessment of current developments in electrochemical sensors using carbon nanomaterials. Carbon nanomaterials are assessed and fabricated particularly focusing the sensor main factors such as sensitivity, selectivity, and limit of detection range. Carbon is one of the promising materials used as electrochemical electrodes because of its unique electrochemical properties such as low cost, low background current, and high surface area [3]. Biocompatibility of carbon nanoparticles is used to analyze the several analytes. The electrochemical analysis technique has been employed to determine the various amounts of electro-active analytes in both qualitative and quantitative terms. These procedures were extremely accurate, dependable, and inexpensive. Various characterization methodologies are widely employed to record the electrochemical response such as pulse amperometry (PA), differential pulse voltammetry (DPV), square wave voltammetry (SWV), and cyclic voltammetry (CV).

Recent surveys summarizes substantial research into nanocomposites based on the coupling of nanomaterials with inherently conducting polymers [4–6]. The diverse application of high yield nanomaterials/conducting polymer-based electrochemical sensors, immunosensors, gas sensors, and biosensors in the fields of drug delivery, microbiology, pesticides, genomics, and other fields has been basically focused,

as well as the future [7, 8]. The schematic diagram for various applications of biosensor is shown in Fig. 2. The electrochemical techniques are very easy to use and handle. So, the more scientific importance is diverted on practical applications like non-invasive point-of-care devices and disposable wearable sensors. Comprehensive review article is exploring the role of carbon nanomaterial as an electrochemical sensor.

### Overviews of carbon-based nanomaterials and graphene

Most recent developments in nanomaterials are based on diverse shapes, sizes, and different compositions for practical employability which includes different metal nanoparticles with different dimensions and hybrid nanocomposites also [9–12]. These nanoparticles are increasingly being employed in testing to increase analytical performance and simplify the detection procedure due to their unique physical and chemical features. They are used as labels for signal creation, transduction, and amplification or as carriers for biorecognition components to be immobilized. ‘Nanotechnology’ can be used to determine disease biomarkers such as cancer cells, enzymes, nucleic acids, viruses, and proteins. Biosensors are widely employed in a variety of applications such as monitoring of glucose in diabetics [12], monitoring of different pollutants in waste water to detect hazardous materials [11], and molecular imaging. 0-D, 1-D, and 2-D carbon-based nanomaterial in conjunction with metal nanoparticles (NPs), enzymes, polymers, DNA, and ionic liquids offer a wide range of biosensor choices.

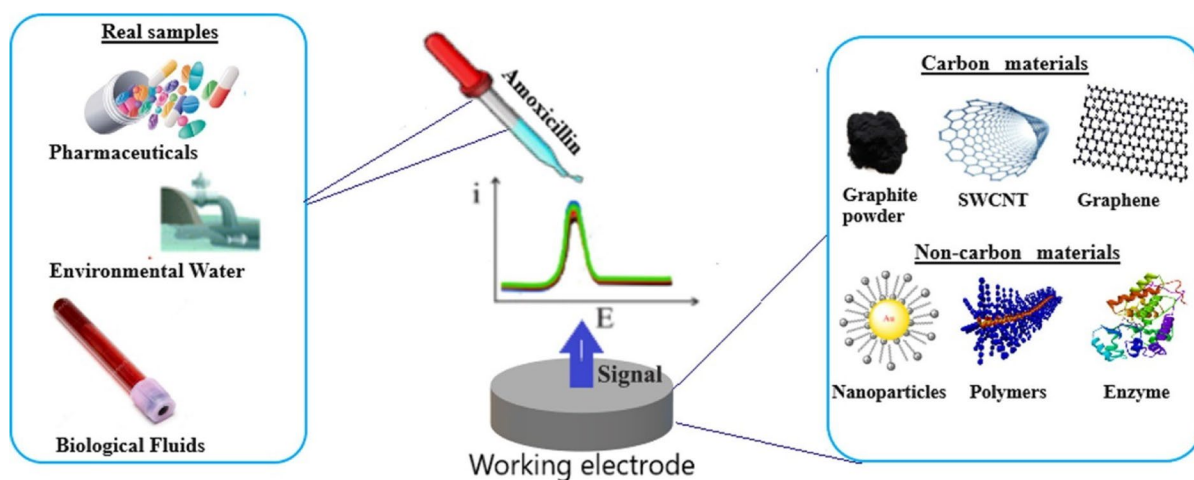
Nanodiamonds, 1D nanotubes, and 2D graphene nanosheets can all be used as prototypes for nanocomposites. ‘Carbon’ atoms with a valence of four can form only one, doubles, and tripled covalent bonds with other elements and each other. Different NP forms include tubes, horns, spheres, and ellipsoids. Carbon nanotubes (CNTs), graphene, rGO, carbon nanodiamonds (CNDs), and carbon dots (CDs) were the maximum critical allotropic adjustments of nanoscale carbon materials [13]. Carbon nanomaterials offer a wide range of technical uses showing improved performance in nanoelectronics, gas sensors, textiles, composites, conductive polymers, batteries, and paints. The benefits such as easy

fabrication, catalyst dispersion, reproducibility, stability, and small ohmic resistance are advantages of carbon-based materials as a working electrode. Due to unique properties of 2-D graphene (GNR), graphene, and its by-products, single and multi-walled carbon nanotubes are the choices to create new foundations for biosensor applications [14, 15] as explored in the following schematic (Fig. 1).

### Manufacturing of electrochemical biosensor exploration

Electrochemical sensor utilization for biochemical sensing has seen tremendous advancements in recent years, resulting in major benefits in variety of applications. In medical applications such as illness diagnostics, the electrochemical sensing technique has proven to be quite useful. The real-time measurement of numerous analytes is possible because of greater sensitive and selective nature of electrochemical sensors. Sensors are the device to detect the very low input signal and able to convert it to different scalable readings. Certain analytes and sample types, as well as other environmental parameters, are commonly employed with these sensors. Electrochemical sensors are utilized in a huge variety of applications, which includes commercial product pleasant control, human fitness interest monitoring, emission prediction, scientific diagnostics, and domestic protection alarms, among others [16–19]. The parametric nature

of sensor devices is used to build electrochemical sensors. In the majority of cases, the characteristics are electronic, with changes in current, voltage, or reactance influenced by analyte configuration. Any type of solid, liquid, or gaseous analytes can be detected with electrochemical sensors. Electrochemical biosensors are also used in a variety of applications such as medical diagnostics and environmental gas monitoring. They are commonly used to monitor the bioactivity in a variety of chemicals and organic materials. Biosensors work by detecting the concentration of a target biomolecule using a biomolecule. Because of the transduction processes used, biosensors are usually classified as a subclass of chemical sensors. Combining both the electrochemical transducers and an electrodes can be used as biomolecule identifiers. The biosensor detects enzymes, cells, antibodies, nucleic acids, and other biological activity using the following concept. The inherent idea is to get the required response amid the base elements and then demonstrate sensitivity with a physical or chemical transducer. The reactive circumstance range obtained by continuous or discrete nature of the signals is the elementary fact for the analysis of biosensor device. Biosensors based on electrochemical are comprised of a delicate filament and an electrochemical signal transducer. The sensor film is utilized to detect analytes, while the electrochemical signal transducer turns biomass activity into an electrical signal. Potentiometric, amperometric, and impedimetric transducers are the three types of electrochemical biosensors based on the nature of

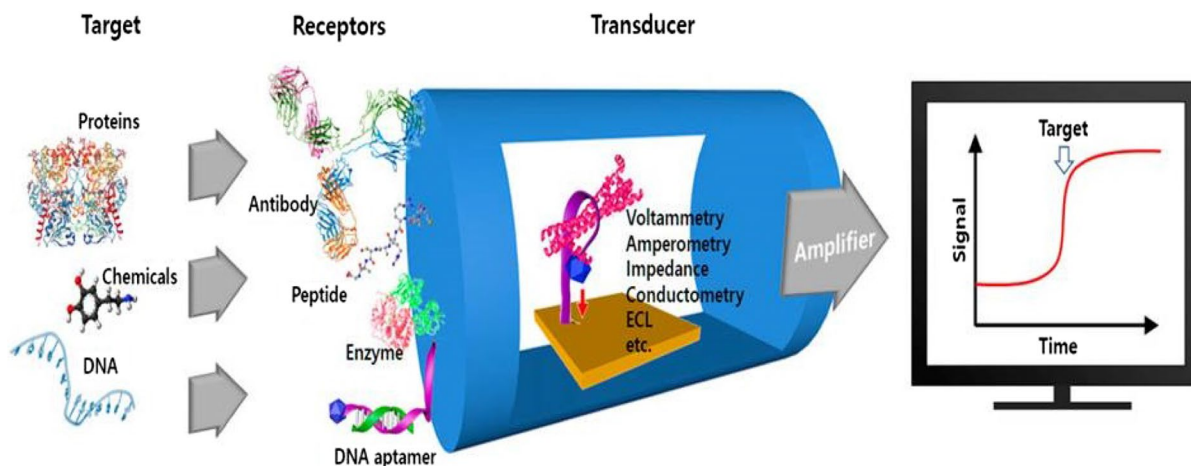


**Fig. 1** Schematic diagram of individual allotropes of CNTs as electrochemical sensors [15] (Copyright © 2021, Elsevier)

the transducer [20–22]. The chemical information is converted into detectable resistive, amperometric, and reactive signals by these separate transducers. The most frequent materials used in electrochemical sensors are electrodes and supportive substrates for electroanalytical characteristics and the identification of biological components. An electrochemical biosensor is depicted in Fig. 2. In this system, biological recognizing devices are attached to the electrodes. The electrode element transducers transform the electrical signal into a readable response that the handling unit can monitor [23–25]. Electrochemical biosensor improvement and adaption seem to have a brilliant destiny in advance of them. Technological developments in electrochemical biosensors offer a variety of real-world applications that have opened a new era in the science of chemistry as depicted in Fig. 2, thanks to an incorporation of specialized biochemical recognition approaches to clinical, drug delivery, cancer treatment, health care, water management, as well as environmental monitoring.

Electrode materials include i.e. platinum (Pt), graphite (GR), gold (Au), carbon paste (CP), and glassy carbon (GC) which may be regularly mixed with nanomaterials consisting of grapheme, carbon nanotubes (CNTs), and derivatives or carbon-based materials. Electrochemical sensors' fundamental purpose is making them valuable in disciplines such as electrocatalysis, electroanalysis, and corrosion, among others. The applied voltage is one of the most useful variables for changing the physical–chemical characteristics of the

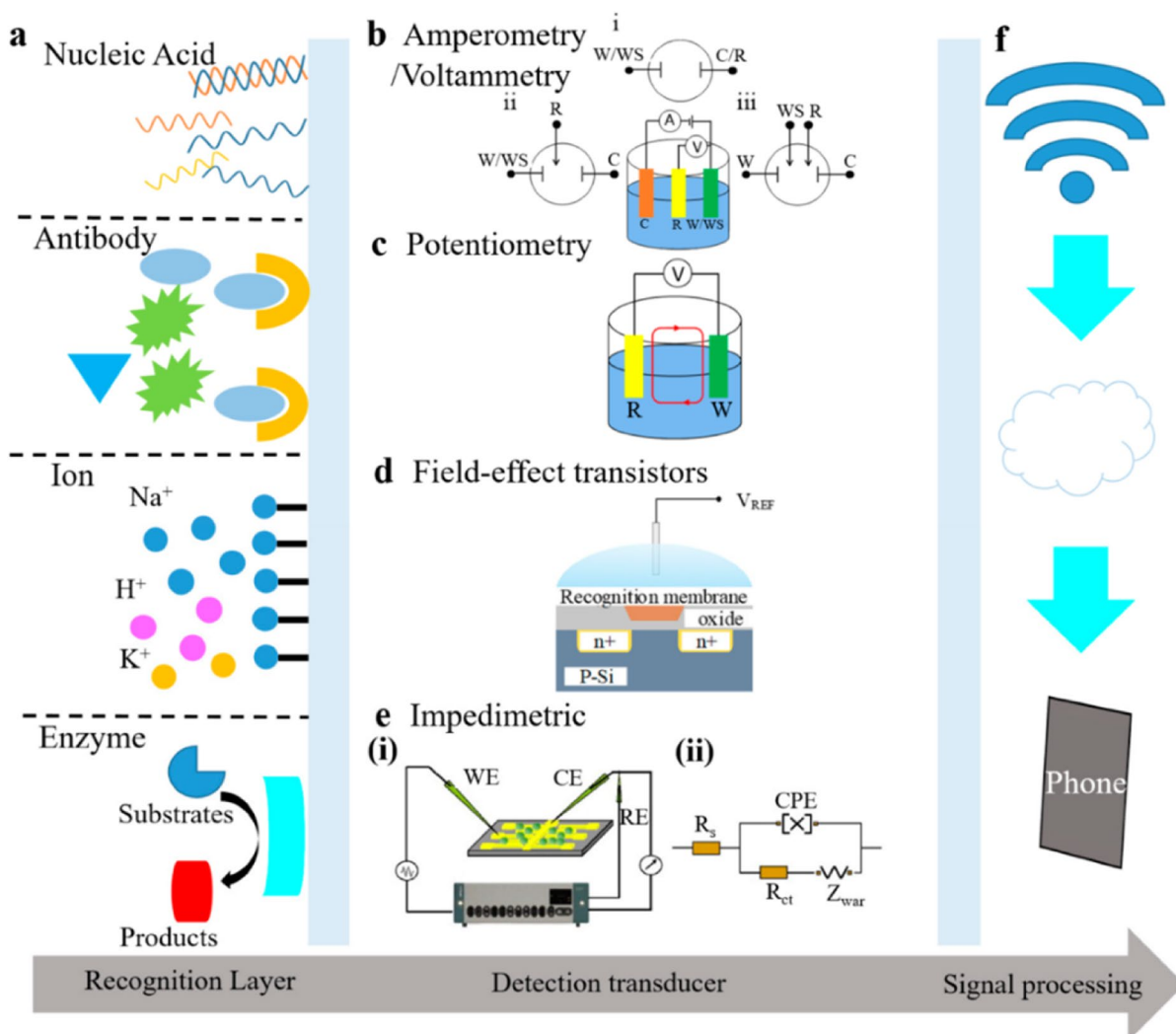
electrode/solution contact in this regard. However, this technique only allows for minor adjustments and has a low selectivity in general. As a result, significant researchers are focusing their efforts on studies involving the intentional adjustment of the electrode surface in order to impose and regulate its features. Chemically active species can be immobilized on the electrode surface to produce a chemically modified electrode (CME). The modified electrode is made up of two parts: a basic electrode (substrate) and a chemical modifier. Because, it needs to have good enough electrochemical traits and compatibility with the immobilization technique selected, the bottom electrode material selected is vital with inside the production of a chemically modified electrode. The use of novel materials/nanomaterial to change different electrode surfaces, as well as the technical uses of these chemically modified electrodes, has increased dramatically in recent years [26, 27]. Although the majority of articles discussed a wide range of materials that can be utilized to modify electrodes, recent years have seen an upward trend towards extruding electrode surfaces with functional nanomaterials, such as graphene, carbon nanotubes, polymers, and metallic oxides. The capacity to carry out an evaluation in situ and in real world, easiness in automation, portability, miniaturization, and occasional price distinguish those sensors from conventional instrumental methods. With their numerous useful properties (such as large surface area, outstanding conductivity), a variety of nanomaterials have been investigated for the discovery of greatly effective electrochemical sensors to



**Fig. 2** Schematic diagram of typical biosensor [25] (Copyright © 2021, Appl. Sci., MDPI)

analyze several target molecules, including neurochemicals like serotonin, hypoxanthine, uric acid, xanthine, dopamine, epinephrine, ascorbic acid, norepinephrine, and acetylcholine. Nanomaterial primarily based totally electrochemical sensors for identifying neurochemicals in frame fluids has a vibrant future. Owing to their excessive selectivity and specificity, they are gaining presently the finest attention. When choosing a species to modify electrodes for electrochemical sensing, great sensitivity, short boundary of detection, and high stability are all desirable characteristics. As electrode

materials, metal oxide-derived structures for the building of electrochemical sensors are discussed with breakthroughs and techniques on metals, metal oxides/hydroxides, sulfides, phosphides, carbons, or their composites as illustrated in Fig. 3. The most essential applications of sensors utilize further recognition element such as nucleic acid, antibody, ions, or enzyme as illustrated in Fig. 3a, while amperometry, potentiometry, field-effect transistors (FETs), electrical impedance analysis of impedimetric, and transduced signals from the sensing element can be transmitted via wireless



**Fig. 3** Electrochemical biosensors utilizing various techniques: **a** a recognition of elements consisting of nucleic acid, antibody, ions, or enzyme; **b** amperometry; **c** potentiometry; **d** field-effect transistors (FETs); **e** electrical impedance analysis

of impedimetric; **f** wireless communication module for analyte detection and concentration analysis [27] (Copyright © 2020, MDPI, 2020)



communication module which is demonstrated in Fig. 3 **b c d e** and **f** respectively.

### Electrochemical operational methodology and detection of CNTs

Researchers have investigated the use of single-wall carbon nanotubes and multi-wall carbon nanotubes as electrodes unmodified or modified for detecting applications. Due to their attractive characteristics, sensors and electrodes based on carbon are frequently deployed in electroanalytical uses owing to its large potential windows, low background currents, variable surface chemistry, and adaptability to diverse sensing tasks [28–30]. The carbon nanotube and the carbon nanofiber are excellent candidates for detecting inorganic and organic chemicals. Nanostructured carbon electrodes should be replaced with silver or copper nanoparticles for specific electroanalytical applications, such as the detection of individual, selective, or simultaneous substances. Nanostructured carbon composite electrodes associated with various electrochemical methods for sensing applications such as voltammetric/ampereometric sensors are the focus of this review.

Cyclic voltammetric method (CV) for critical analysis

The electrochemical characteristics of the electrode surface, combined with certain electrochemical procedures, determine the electrode's detection performance. Cyclic voltammetry (CV) is used to investigate the electrochemical characteristics of the electro-active

surface area (ESA). To determine the properties and processes of a redox system, electrochemical techniques are employed. This is an introductory electrochemical test used to illustrate an electrode substantial to use in a variety of applications [30]. Besides revealing thermodynamic redox processes, this technique can also reveal the kinetics of heterogeneous electron transfer reactions and associated chemical responses. As a working electrode is compared with a reference electrode, a counter electrode is observed in a supporting electrolyte. It is well well-matched for detecting redox couples quickly in a system. Through a more detailed study of the cyclic voltammogram, a couple can be identified. Using a triangle potential waveform, potentials are scanned backward and forwards linearly with time. Electro-active surface area is commonly determined by the ferro/ferricyanide redox couple technique. The features of a cyclic voltammetry reaction caused by a reversible procedure can be determined by recording cyclic voltammetry at different scan rates in the presence of 4 mM  $K_3Fe(CN)_6$ , as investigated in the graphical analysis in Fig. 4.

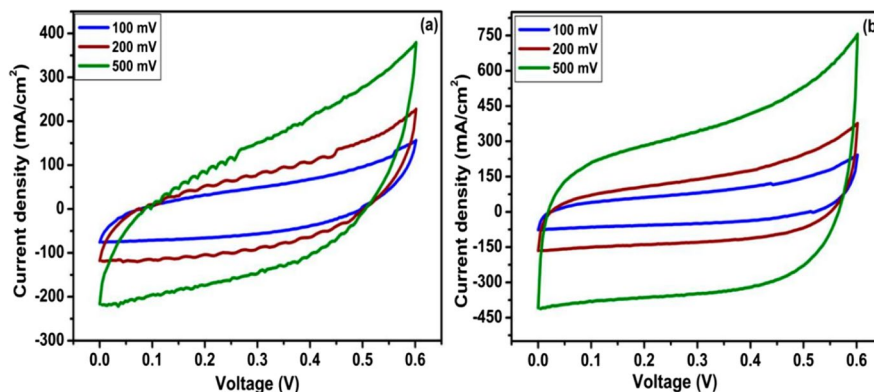
Differential-pulsed voltammetry

The differential-pulsed voltammetry (DPV) technique is highly sensitive to low levels of analytes. The DPVs are illustrated in Fig. 5. The differential pulse voltammogram height directly relates to the concentration of the materials are utilized.

Square-wave voltammetry (SWV) analysis

Figure 6 shows SWV is a high amplitude differential approach with greater sensitivity than

**Fig. 4** Cyclic voltammetric curves of **a** titanium dioxide CNTs and **b** nitrogen-doped  $TiO_2$  nanotubes [28] (Copyright © 2019, MDPI)



DPV, particularly for reversible systems where the reverse pulse causes the reverse response, and the forward and reverse components differences are recorded.  $f\Delta E$ , where  $f$  is frequency and  $E$  is the step potential, gives the effective scan rate (SP).

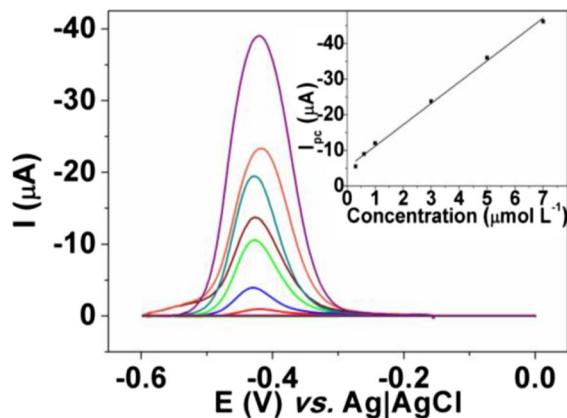
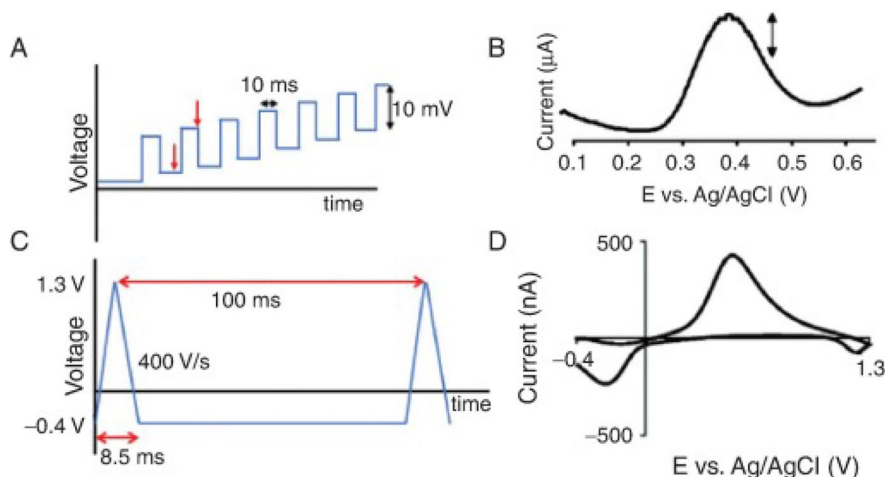
### Chronoamperometry analysis

The simplest basic electrochemical recognition method, chronoamperometry, is adequate for the majority of real-world uses. This method's current–time dependency is monitored at a constant potential value, which is chosen based on cyclic voltammograms' well-known vital theme of reference. Because diffusion regulates mass transport, changes in the concentration gradient near the electrode surface are reflected in the current–time dependency.

### Detection strategy of pulsed amperometry

Pulsed amperometry is a substitute to chronoamperometry that eliminates the drawbacks of electrode fouling in the detection applications. In pulsed amperometric detection programs, the measurement capability is held for a small period (measurement pulse) after the electrode cleaning and conditioning pulses. In situ cleaning and reactivation of the electrode surface during an electroanalytical method of finding has proven to be particularly sensitive for the identification of organic compounds. The PA method is chiefly useful for detecting analytes that are easily

**Fig. 5** Differential pulsed voltammograms [29] (Copyright © 2020, Elsevier)



**Fig. 6** Square wave voltammetry (SWV) obtained under optimized conditions in a  $0.04 \text{ mol}\cdot\text{L}^{-1}$  B-R buffer solution with increasing concentrations of AG25: (a) blank, (b)  $1.0 \times 10^{-7}$ , (c)  $5 \times 10^{-7}$ , (d)  $1 \times 10^{-6}$ , (e)  $3 \times 10^{-6}$ , (f)  $5 \times 10^{-6}$ , and (g) to  $7.0 \times 10^{-6} \text{ mol}\cdot\text{L}^{-1}$  [30] (Copyright © 2015,MDPI)

adsorbed on the electrode surface in practical detection applications.

### Carbon nanotubes (CNTs) uses for electrochemical biosensors

One of the maximum fast-growing clinical subjects is the observation of the electrochemical pastime of bioactive compounds. The use of carbon-based nanomaterials in operational electrodes, such as nanomaterials, 2D graphene, and graphene oxide, is a quickly

developing field of biosensor design. Electroanalytical strategies are cost-effective, handy use, accuracy, and reliability benefits over different detection strategies including chromatography, luminescence, and spectroscopic strategies. The electrochemical study is a straightforward and cost-effective approach for determining the amounts of electro-active species in a solution, both quantitatively and measurably.

Researchers can use a variety of methodologies to investigate the electrochemistry of electro-active substances in solution. Differential pulse voltammetry (DPV), cyclic voltammetry (CV), and linear sweep voltammetry (LSV) are the most used electrochemical sensor detection methods [33, 34]. After optimization, for the greatest electrochemical response, they are all useful electroanalytical procedures. The situation of the analytes under examination, the category of electrode utilized, and the electrolyte used can all influence these processes. The voltammetry response of the system can be influenced by the dimension and morphology of the electrode, as well as the fabrication technique deployed. The chemical and physical qualities of electrode surfaces, adsorption, the result of the applied voltage, and coatings added to the electrode surface to increase detection all play a role in electroanalysis methods. Carbon compounds are commonly exploited in electroanalytical studies due to their chemical inertness, small background current, quite extensive potential range, and adaptability to many types of analysis. Figure 3 depicts the detection of metabolic diseases by carbon nanomaterial based on electrochemical sensor. Detection of metabolic disorders by electrochemical biosensors has gained a lot of recognition in current era. As a result, the current research and development of various carbon nanomaterials to detect electrochemical biosensor of general metabolic disorders markers is discussed in this paper.

A carbon nanotube (CNT) is an allotropic modification of carbon. Carbon atoms with three electrons create s-bonds trigonally synchronized to three carbon atoms in CNTs through  $sp^2$  hybridization. One layer of graphene is rolled into a smooth hollow tube to form CNTs. Rolls of graphene sheets arranged into cylindrical arrangements or tubular arrangements having diameter of quite a few nanometers make up carbon nanotubes. CNTs can be customized in diameter, length, number of layers, and chirality vectors (symmetry of the nailed graphite sheet). A number of researchers are inclined to carbon nanotubes (CNTs), which have

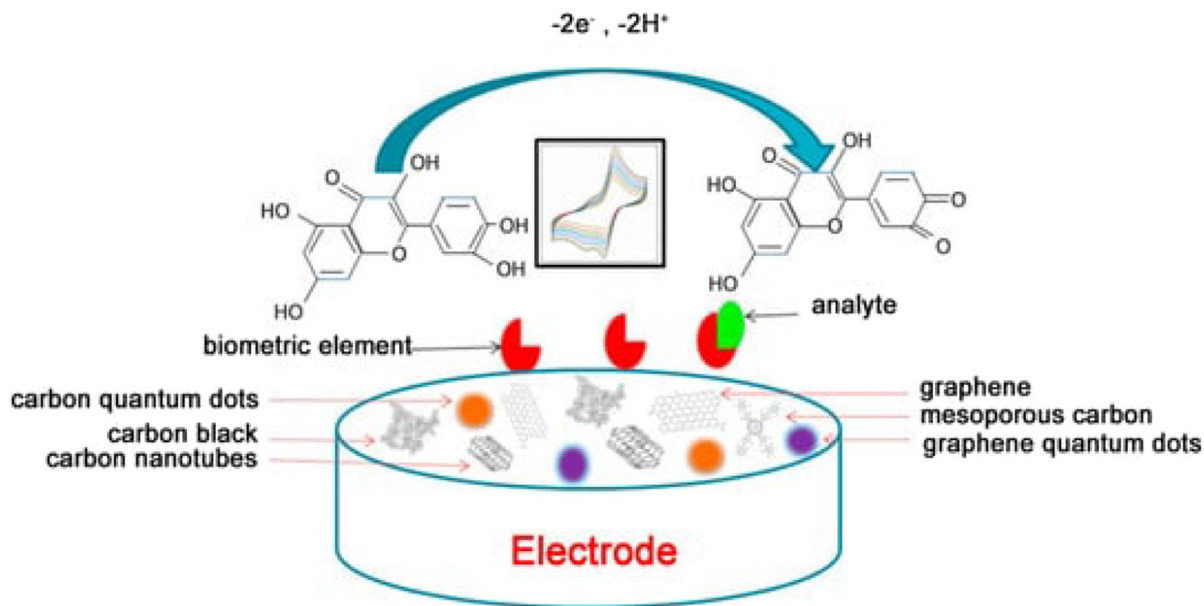
$sp^2$  hybridized carbon bonds and contain multi-walled carbon nanotubes (MWCNTs) and single-walled carbon nanotubes (SWCNTs), due to their interesting shapes [35–37]. MWCNTs process a diameter from 5 to 25 nm and a length of about 10 m, whereas SWCNTs process a diameter from 1 to 3 nm and a length of a few micrometers. CNTs possess superior physical properties, such as stiffness, strength, and elasticity, when compared to other fibrous materials. Unlike other materials, CNTs have a higher aspect ratio (length to diameter).

These nanotubes are formed from a single or multilayer graphene sheet curled at a specific helical angle around a central axis. Due to their extensive range of structural, thermal, and electrical characteristics, they have been used as catalyst materials. Nanocarbon has a large specific surface area, excellent conductivity, and is easy to modify. CNTs are exploited in electrochemical reactions as electrodes, and they handover electrons more efficiently. Additionally, they have a lot of potential as biosensors because they can immobilize proteins while maintaining their intrinsic properties. As shown in Fig. 7, CNTs have been deployed as electrode material in a variety of electrochemical sensors owing to their capacity to enhance electron transfer processes with electro-active species in solution and at the electrode interface. As a result of their superior conductivity and chemical stability, CNTs have excelled in materials previously used as electrode interfaces.

#### Carbon-based hybrid nanomaterial synthetic approach

Carbon is an incredibly adaptable material with a variety of unique properties. From 1-D to 3-D structures, it is available in a variety of allotropes and is used for a number of applications. Many of the essential features of the various allotropes have been fully defined in the recent studies on the categorization and uses of single carbon allotropes. True carbon-primarily based totally hybrid nanomaterials are described as a brand new smart electronic material wherein or extra carbon allotropes were incorporated into a brand new hybrid with feasible additions of certain metal nanoparticles and which reveals rising properties which are drastically past the ones of its constructing blocks. Furthermore, the hybrid's fabrication technique should be repeatable and controllable, permitting technique scaling and tool shrinking.





**Fig. 7** Carbon nanotubes used in electrochemical sensors as an electrode material [31] (Copyright © 2020, MDPI)

A true hybrid material is an arrangement with a diamond-like carbon (DLC) thin film as a functional substrate, 'Ni' metal placed on top of that, and carbon nano films developed on top of that [32–38]. As a result, a carbon-primarily based totally hybrid nano-material is formed, with carbon nano film with 'Ni' particles at their tips, and the DLC layer modifies the carbon nano film form from tubular to platelet-like for the duration of the early levels of increase via way of means of performing as an additional carbon source. In this manner, the CNF arrangement is strongly fused with the substrate, guaranteeing a strong mechanical and electrical bonding. These carbon-based hybrid nanomaterials can also be used to manufacture electronics because DLC can be patterned and is CMOS compatible. Combining allotropic nanocarbons like fullerenes, graphene, carbon nanotubes, carbon nano buds, and nanodiamond brings up systematic and scientific potentials that no other single element or substance can equal. Nanocarbon compounds, for example, can act as a practical interface among the residing and non-residing worlds. (i) They are expected to be game-changing materials in the field of biomedical engineering as a result. (ii) Second, carbon will never become a critical substance because it is abundant in nature. (iii) Finally, nanocarbons thermal, electrical, electrochemical, optical, biological, and different

physicochemical homes are not most effectively use in and of themselves; however, nanocarbon-primarily based totally hybrid substances may be produced to acquire the preferred blend of capabilities [39, 40].

#### Basic synthesis of "Graphene"

A thin layer of carbon atoms in a 2D allotropic form of carbon is known as graphene. Carbon atoms form a hexagonal crystal lattice with an interatomic spacing of 0.142 nm in  $sp^2$  hybridization, which is connected by sigma and 'pi' bonds.

Graphene is a semiconductor material having zero band gaps, and ambipolar electric field and charge carrier mobility of more than 15,000 to 20,000  $cm^2 V s^{-1}$  at room temperature. It is 97.7% light translucent and has notable physical, thermal, mechanical, and chemical characteristics [41, 42]. When as compared to ordinary noble metals which include Au, Ag, Cu, Cr, and Al, graphene well-known shows a decrease in power losses which include Ohmic and irradiative losses, in addition to excessive tunability. As a result, it is a usable candidate for electrochemical sensors with great sensitivity.

Electron mobility in graphene layers is 100 times greater than in silicon layers. Graphene is widely used

in sensors because to its greater charge carrier mobility and huge specific surface area. Graphene-modified electrodes and devices for use in electrochemical sensors have been created using the CVD method confirmation of structural morphology via FE-SEM in Fig. 8a,b and HR-TEM illustrated in Fig. 8c,d. Graphene has a huge specific surface area ( $> 2500 \text{ m}^2 \text{ g}^{-1}$ ), a high thermal conductivity ( $> 3000 \text{ W mK}^{-1}$ ), and a substantial Young's modulus ( $> 0.5\text{--}1 \text{ TPA}$ ). It also has a large specific surface area ( $> 2500 \text{ m}^2$ ), which can create robust connections with biomolecules, outstanding transparency, electron conductivity, and greater mobility ( $> 2 \times 10^5 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  at electron density  $2 \times 10^{11} \text{ cm}^{-2}$ ) and excellent transparency, electron conductivity, and superior mobility ( $> 2 \times 10^5 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  at electron density  $2 \times 10^{11} \text{ cm}^{-2}$ ). As a result of carbon's  $\text{sp}^2$  hybridization, graphene refers to a single layer of carbon atoms structured in a 2D crystalline hexagonal lattice [43, 44].

As a result, graphene contains strong in-plane sigma connections, which explain its mechanical strength and flexibility, as well as weak out-of-plane pi bonds, which explain its thermal conductivity, electrical charge, and transparency. By folding in different ways or aggregating some layers, graphene as a fundamental material can produce several carbon allotropes, namely 0D fullerene, 1D CNTs, 2D graphene,

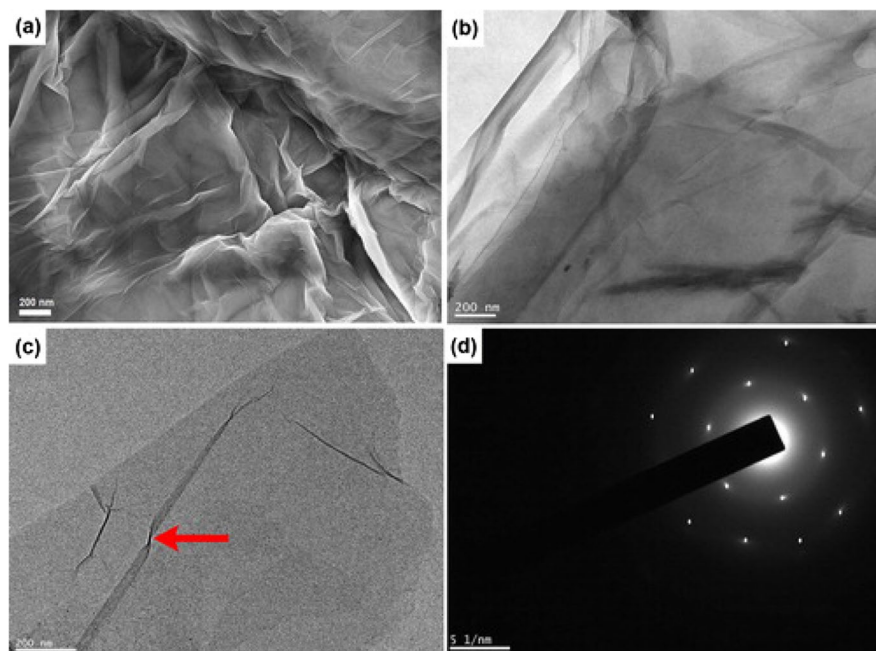
and 3D graphite [45–48]. Monolayer and multilayer graphene, graphene oxide, reduced graphene oxides, and graphene quantum dots are all members of the graphene family.

Graphene and its end product offer certain distinctive physicochemical feature, such as a huge surface area of  $2630 \text{ m}^2 \text{ g}^{-1}$ , easiness in solubility, large drug loading capability, good purity, and cell membrane infiltration flexibility. Graphene has a variety of uses, including apparent conductive sheets, nanoelectronics, and touch displays. It can be manufactured by CVD or repeated mechanical exfoliation, but defect-free production is difficult. Monolayer graphene can be employed in biosensor design because of its outstanding mechanical strength and thermal conductivity, as well as a tuneable electrical band gap.

#### Chemical approach to synthesis of graphene oxide (GO)

Graphene oxide (GO) is an oxidized variant of graphene and is a single-atomic layered polymer generated by extreme oxidation of graphite. Graphene oxide (GO) has been created at a very low cost by chemical oxidation of graphite [50, 51]. For the growth of GO, the Hummers method was used because it takes very little time and does not dissolve

**Fig. 8** Typical microscopic illustration of **a** FE-SEM image of reduced graphene oxide (rGO) at magnification 200 nm resolution, **b** HR-TEM image captured at 200 nm resolution, **c** single layer crystalline patterns graphene oxide as shown by “red” arrow, and **d** selected area diffraction (SEAD) patterns shows single crystallinity of graphene oxide [40] (Copyright © 2021, Taylor & Francis)



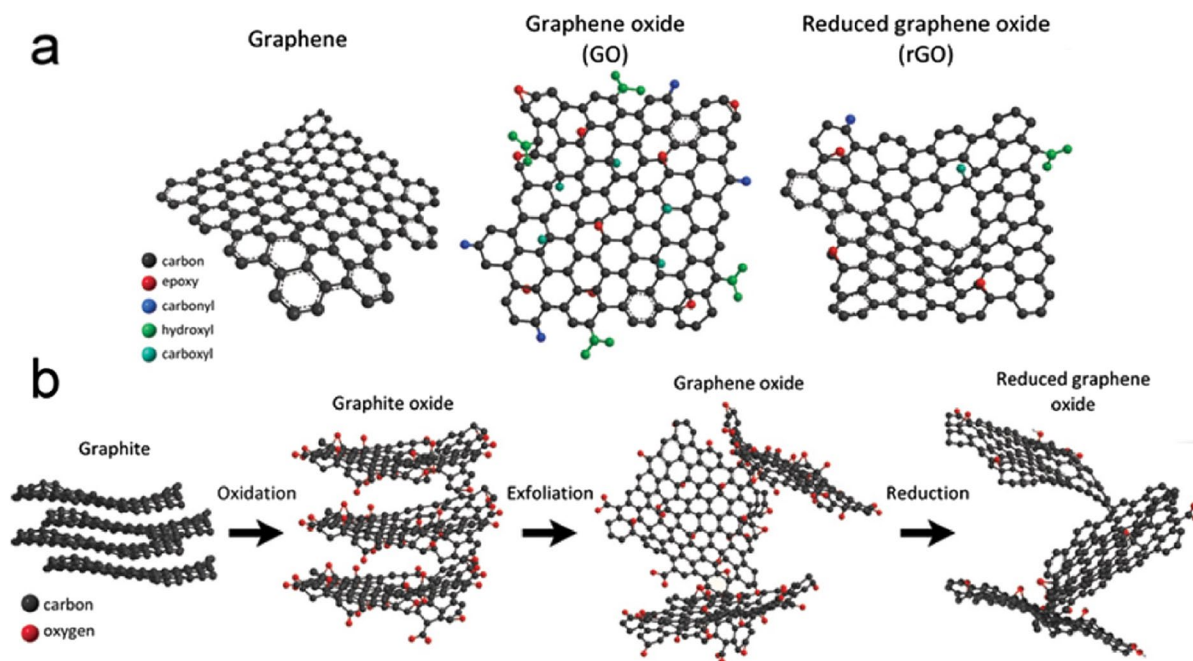
hazardous compounds. The use of  $\text{KMnO}_4$  and concentrated  $\text{H}_2\text{SO}_4$  as an oxidation agent and for peeling off graphite has also been described in the manufacture of graphene oxide (GO). The hydrophilic nature of GO is due to the acidic treatment.

The GO sheets are easily diffused in water. Chemical reduction procedures, namely the direct adding of reducing agents like hydrazine or thermal reduction at large temperatures, can be used to decrease GO back to graphene. Epoxy ( $>\text{O}$ ), hydroxyl ( $\text{dOH}$ ), carbonyl ( $\text{CO}$ ), and carboxylic ( $\text{dCOOH}$ ) groups are among the oxygen-containing functional groups found in GO. GO is water (and other solvents) dispersible, which is schematically represented in Fig. 9. The  $\text{sp}^3$ -hybridized carbon and oxygen functional groups are exposed in both amorphous and crystalline defect areas of GO. The GO's strong water solubility, oxygen-containing groups, high adsorption capacity, superior biocompatibility, and outstanding stability make it ideal for immobilizing proteins, nucleic acids, and other biomolecules as thin films [52–54].

Chemical growth dynamics of graphene has been described to develop GO, in which graphite is

oxidized, resulting in an aqueous colloidal form of GO flakes. As a consequence, hydrophilic functional groups are functionalized in the graphene's basal plane. GO has been found to be compatible with single-strain DNA (ssDNA), peptides, and amino acids in several investigations [56, 57]. The oxygen functional groups in GO can be decreased via thermal, chemical, and electrochemical processes to generate reduced GO (rGO). The number of oxygen functional groups in rGO is lower than in GO.

Reduced graphene can be thought of as a bridge between pristine graphene and heavily oxidized graphene, keeping some but losing some of the features of both materials [58]. On GO, their interlayer distance was  $7.9 \text{ \AA}$ , whereas, on rGO, it was  $3.4 \text{ \AA}$ . This material can be used in biological and biosensor applications by adjusting the carbon-to-oxygen ratio and chemical contents in rGO. Furthermore, GO is an appealing nanocarrier for cargo delivery due to its easy and efficient cell penetrance as well as its ability to preserve DNA and peptides from enzymatic cleavage [59]. Furthermore, GO's unique optical and electric properties, such as fluorescence quenching efficiency, in combination with its biofunctionality,



**Fig. 9** (a) Structure (Chemical) of graphene (G), graphene oxide (GO) and reduced graphene oxide (rGO) (b) synthesis route of graphene (G) to reduced graphene oxide (rGO) (Copyright © 2020, MDPI) [64]

made it an ideal choice for biosensing and bio imaging applications [60]. Controlling the oxidation amount of GO allows you to modify optical transparency, as well as electrical and mechanical properties. In comparison to substantially reduced GO, accumulative oxidation would surge the quantity of oxygen-containing groups, diminishing its electric capabilities [61].

GO has also been proven to have antimicrobial properties. The optimal smart characteristics of GO is that it could be partly minimized to graphene-like materials by eliminating the oxygen-containing clusters, regaining some of graphene's structure and characteristics. The ultimate goal of decreasing GO is to create graphene-like sheets that are structurally and functionally identical to pristine graphene made from graphite. Despite many efforts, it is difficult to attain this goal since the remaining functional groups and structural flaws significantly alter the properties and structure of carbons.

As a result, even for the sake of simplicity, we are not permitted to refer to rGO as graphene. This would open up a promising path for graphene production and applications on a large scale. The chemical and thermal reduction strategies use various reducing agents, namely hydrazine, which is extremely poisonous and also creates GO with nitrogen heteroatoms. NaBH<sub>4</sub>, vitamin C, and hydrophilic acids are all alternatives to hydrazine, and rGO in blend with nanoparticles and polymers offers an extensive series of applications in tissue engineering, disease biosensing, cell culturing, and eliminating ecological contaminants such as nitroaromatics. For the loading of a variety of therapies, such as anticancer medications, weak soluble pharmaceuticals, antibiotics, peptides, DNA, RNA, antibodies, and genes, GO may offer benefits over other nanomaterials [62–64].

#### Synthesis of r-GO (reduced graphene oxide)

It is a type of graphene oxide that has been reduced in size. Reduced graphene oxide was also obtained using the electrochemical reduction approach (rGO). Various reduction methods, including laser radiation, annealing, and chemical procedures, have been used to partially reduce Go to create reduced graphene oxide (rGO) [65, 66]. The use of severe chemicals for oxidation, on the other hand, impairs the characteristics of graphene by destroying the basal plane. That is why graphene is separated from graphite using appropriate

solvents and surfactants. In some liquids, graphene tends to clump and form graphite. Preparing pure and consistently distributed single-layer graphene in solvents is thus difficult and challenging. Graphite is mechanically peeled to obtain pure 2-D graphene, which is achieved by using adhesive tapes with a lower defect density [59, 60].

#### Fullerenes in electrochemical biosensor applications

Fullerene, an allotropic change of carbon, turned into observed in 1985. It turned into the primary nanomaterials to be separated successfully. Fullerene derivatives were proven to be powerful withinside the photoacoustic imaging of most cancers and tumor cells. The production of a number of atomic C<sub>n</sub> clusters ( $n > 20$ ) of carbon atoms on a spherical surface is a distinguishing property of fullerenes. In fullerenes, the carbon atoms establish covalent bonds with one another during sp<sup>2</sup> hybridization [67, 68]. They are most typically found at the apexes of hexagons and pentagons on the sphere's surface. In the medical field, fullerenes have been employed in cancer treatments, MRI, and gynecological diseases. Fullerenes are a 0D form of graphitic carbon that resembles an uneven sheet of graphene wrapped up into a sphere by addition of pentagons to its structure.

They arise in a variety of shapes and sizes, with carbon atom counts ranging from 30 to 300. Sputtering, electric arc discharge and electron beam ablation are all processes that can be used to make them [69, 70]. Fullerenes can also be manufactured using graphitic electrodes and can be found in the soot of combustion flames. The first fullerenes were made by evaporating graphite electrodes in a helium environment. However, due to the high cost of synthesis and low yields of the methods, fullerenes have limited practical application. Nanomaterials are commonly used in the manufacture of electrochemical biosensors to expand analytical performance as they may enhance the surface area, boost electrocatalytic activity, and expedite electron transfer to electrodes. CNTs, Fe<sub>3</sub>O<sub>4</sub> particles, and AuNPs are examples of single nanomaterials, while hybrid nanocomposites containing several nanomaterials, namely the blend of CNTs and AuNPs, are examples of hybrid nanocomposites.

These biosensors are used in a variety of nanomaterials, including quantum dots, magnetic nanoparticles, CNTs, fullerene, and graphene oxide.



A biological component such as microorganisms, enzymes, organelles, tissues, antibodies, and cells is utilized in the identification site of a biosensor. A range of analytes, including proteins, enzymes, and nucleic acids, have been detected using electrochemical biosensors, and fullerene is employed as a facilitator between the detection site and biosensor's electrode to increase the electron transfer rate generated by biochemical or the biocatalytic reaction of analyte in connection with the biological component at the detection site [71, 72].

Fullerene has a completely distinctive topological provenance and electrochemical characteristics, together with wide UV Vis absorption, photothermal consequence, physical angle strain, the capacity to house a couple of electrons and endohedral metallic atoms, long-dwelling triplet state, singlet oxygen production, and the capacity to behave as an electron acceptor with a twin nature of electrophilic and nucleophilic nature. However, elemental fullerene is hydrophobic and insoluble in polar solvents, making bioconjugation with photosensitive and physiologically active molecules like DNA, porphyrins, proteins, ferrocenes, dendrimers, and different compounds challenging.

Fullerene has been efficaciously used in the improvement of biosensors to detect several biomolecules, along with glucose, urea, and proteins, in addition to doping agents, along with dexamethasone, prednisolone, and others in actual samples, ensuing in capability applications. The use is not only confined

to these locations; it also includes a high likelihood of detecting cancer cells at an early stage [73, 74]. In biosensing devices, fullerene acts as a mediator. A biosensor should have two characteristics: it should be hydrophilic and possess several active functional groups that allow it to conjugate with targeted proteins and build a link amid the electrode surface and the recognition site, allowing electrons generated by biochemical reactions at the recognition site to be transferred more efficiently [75–77].

A transducer and a recognition site are also included in biosensors [78–80]. The transducer translates this reaction into a new type of energy which is amplified, processed, and turned into the desired format of signal (Fig. 10). Biosensors come in a different shapes and dimensions, subject to the application, but they all have the same basic configuration.

Carbon nanocomposite utilization for electrochemical sensors as glucose detection

Carbon nanotube fibers made by CVD were employed as a sensing electrode for detection of glucose content. The carbon nanotube-based biosensors responded to the presence of glucose with a very quick amperometric response. Zhu et al. studied and reported extensively on glucose-based CNTs [79, 80]. A glucose biosensor was made using the phase separation method and multi-walled carbon nanotube-grafted chitosan (CS)-nanowire (NW) to

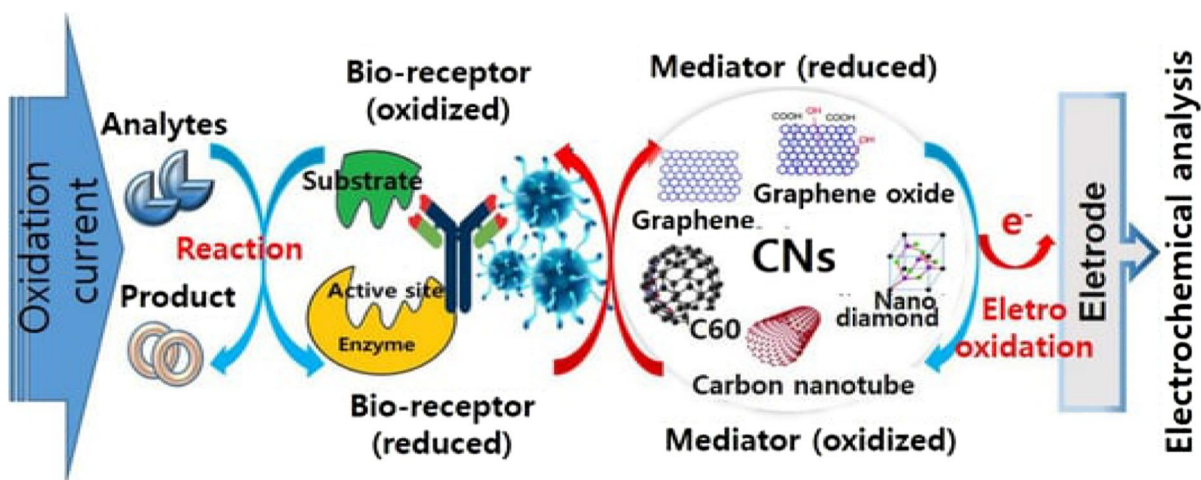


Fig. 10 Components and the involved mechanism of a conventional biosensor [78] (Copyright © 2021, MDPI)



which glucose oxidase was linked to creating the biosensor [81]. Using cyclic voltammetry and amperometry, researchers were able to detect glucose electrochemically.

The constructed biosensor detected glucose with a large sensitivity of 5.03 A/mM in a concentration range of 1–100 mM and a short reaction period. The multi-walled carbon nanotube-chitosan helps glucose oxidase and target molecules conduct electrons more efficiently. Glucose oxidase was covalently connected to CNT nanoelectrode ensembles by forming amide bonds amid the carboxylic acid groups and their amine residues existing on the tips of CNTs in another investigation by M. K. Kim et al. [82]. A biosensor using Ni-nanoparticles dispersed in vertically aligned CNTs grown on Si/SiO<sub>2</sub> substrate was described in another study, with the Ni nanoparticles depositing uniformly inside and on top of the CNT forest. The constructed biosensor had a sensitivity of 1433 A m M<sup>-1</sup> cm<sup>-2</sup> and a detection limit of 2 M over a linear range of 5 to 7 M studied by Z. Zhu et al. [83]. CuO nanoparticles deposited on the sidewalls and tips of vertically well-aligned MWCNTs arrays were studied in another study using a two-step electrodeposition process [84].

Lactate level monitoring is used in sports medicine, biotechnology, and food processing. CNT and mineral oil paste containing lactate oxidase were used to perform amperometric lactate detection. The electronic structures of SWCNTs have an impact on CNTs enzymatic biosensor detection. When connected to metallic SWCNTs, the [Fe–Fe]-hydrogenase enzyme from *Clostridium acetobutylicum* showed improved electrocatalytic activity. This is due to increased coordination between the redox-active sites of *Clostridium acetobutylicum* and the electron surface.

Covalent bonding was used to attach glutamate hydrogenase to the top of CNTs, resulting in a glutamate biosensor with detection limit up to 10 nM. The CNT electrodes are a preferable choice for enzymatic biosensors with excellent sensitivity and selectivity. Polysulfone (PSF) was dissolved in dichloromethane and deposited on a GCE to make the sensor. The PSF layer is drop coated with nitric acid-functionalized MWCNTs. After cross-linking with glutaraldehyde, the tyrosine enzyme (TyOx) is deposited on MWCNT/PSF/GCE. The CV and electric impedance spectroscopy techniques were used to characterize the biosensor [85, 86].

To detect tyrosine, the biosensor has an extremely low detection limit of 0.3 nM and very high sensitivity of 1.988 A M<sup>-1</sup> cm<sup>-2</sup>. The impact of nitrogen doping on the electrochemical overall performance of carbon nanotubes for the detection of H<sub>2</sub>O<sub>2</sub> has been hired to construct a lower-ability H<sub>2</sub>O<sub>2</sub>-primarily based totally enzymatic biosensor mentioned with the aid of using X. Xu et al. [87]. The hydrogen peroxide biosensors have been made using the enzyme horseradish peroxidase (HRP), which becomes related to the surface of MWCNTs the usage of the mediator methylene blue and a cross-linkage among horseradish peroxidase and bovine serum albumin (BSA) composite movie mentioned with the aid of using Q Shi et al. [88]. CNTs changed with Pt nanoparticles synthesized with the aid of using chemical reduction method at the surface of a waxed graphite electrode were defined as a hydrogen peroxide detector. For the detection of glucose existence, glucose oxidase was linked to the sidewalls of SWCNTs. When glucose was added to the glucose oxidase attached SWCNTs, the conductance increased, acting as a biosensor for the enzyme activity.

#### *Ascorbic acid detection via carbon nanomaterials and graphene nanocomposites through electrochemical sensors*

Because of their great architectures, scalable manufacture, and awesome characteristics, carbon nanoparticles are a few of the maximum broadly hired active substances in electrochemical applications [89, 90]. Single-wall CNT (SWCNT), multi-wall CNT, carbon nanohorns (CNH), carbon nanodots, graphene derivatives, carbon nanofibers (CNF), carbon nanoparticles, and nitrogen-doped carbon substances have all been hired as electrode substances for numerous sensing applications. Tashkhourian et al. supplied carbon nanotubes decorated silver nanoparticles composite changed carbon paste electrode (CPE) for the specific detection of AA. With a limit of detection of 12 M, the sensor confirmed linearity from 0.3 to 2000 mM. MWCNT, ionic liquid (IL), and Pd nanoparticles were used to alter any other ascorbic acid sensor primarily based totally on the carbon paste electrode.

A diagram depicting various CNT and graphene additions to composites, as well as six crucial characteristics in CNT and graphene composites, that are expected to improve performance for future

applications is shown in Fig. 5. The detection of AA has been effective using a variety of carbon nanostructures. Xu et al. described the AA sensor with modified GCE with single-wall carbon nanohorns (SWCNH) [90–93]. At room temperature, the SWCNH was produced using the CO<sub>2</sub> laser ablation process. With a small supply voltage, the SWCNH-modified GCE displayed amazing sensor properties. The SWCNH/GCE sensor was used to find the ascorbic acid in genuine samples (serum and medicines). Similarly, the MWCNT reformed electrode to determine ascorbic acid was built positively. The sensor had a high sensitivity of 1287.3 A mM<sup>-1</sup> cm<sup>2</sup> for a linear scale of 100–600 M and a LOD of 7.1 M, according to electrochemical experiments.

Because of the higher electro-active surface area and good conductivity of MWCNT, the PdNP/IL-MWCNT modified electrode outperformed the CPE and PdNP/CPE electrodes in catalytic activity. With the help of conventional addition technique, the sensor was used to measure the concentration of AA in human serum and urine samples. The sensor had great sensitivity; however, it only had a linearity of 112 M. As electrode-surface modifiers, bimetallic nanoparticles (Cu<sub>2</sub>O and Ag<sub>2</sub>O) anchored to functionalize MWCNT were employed in another investigation. Amperometric was used to deposit Cu<sub>2</sub>O and Ag<sub>2</sub>O nanoparticles on an MWCNT-modified Cu substrate.

Graphene, an atomic-scale thick carbon sheet with a zero-band gap, has achieved significant advances in material science and electrochemistry [94]. Previously, it has been deployed as active catalytic material in biosensing, super capacitors, lithium-ion batteries, fuel cells, and solar cells owing to its outstanding electron mobility (200,000 cm<sup>-2</sup> V<sup>-1</sup> s<sup>-1</sup>), chemical stability, greater surface area (2630 m<sup>2</sup>/g), good mechanical strength, outstanding thermal conductivity (5300 W m<sup>-1</sup> K<sup>-1</sup>) and electric conductivity [95]. The selectivity and accuracy of pyrolysis photoresist films (PPFs) modified with graphene nano-sheets based on new ascorbic acid sensors, for example, were confirmed using commercial vitamin C supplements.

The graphene-doped CPE had better sensor qualities than the unmodified CPE in terms of sensitivity, repeatability, long-standing stability, and extensive linearity. The oxidation potential of graphene-doped carbon paste electrode moved to the negative side as compared to unmodified carbon paste electrode [96].

Yang et al. created an electrochemically reduced graphene oxide (ErGO), which they used to detect AA, dopamine, and uric acid [97]. When compared to bare GCE, the ErGO-modified GCE demonstrated exceptional electrocatalytic activity with a little oxidation capability and an eight-fold greater current responsiveness. With a LOD of 250 M, the sensor had a linear detection range of 500 M to 2 mM. An extremely sensitive AA sensor based on pure graphene was also reported. The organic salt-assisted exfoliation process is used to create pure graphene. The sensor demonstrated superior sensing characteristics when compared to graphene that has been chemically transformed (CCG). In comparison to CCG (1250 Sm<sup>-1</sup>) and thermally reduced graphene oxide (600 and 450 Sm<sup>-1</sup>), exfoliated graphene had a huge conductivity (13,000 Sm<sup>-1</sup>).

In comparison to the bare GCE and graphene/GCE, the nanocomposite-modified electrode had a better current response. In addition, Au nanostructures have been used in sensitive electrochemical ascorbic acid sensors. From a research work, a simple electrochemical approach was used to produce the Au nanoplate-decorated graphene hybrid nanomaterials (rGO/Au), which were then used to detect ascorbic acid in spiked serum samples [98]. Small detection capability, a little detection limit and anti-interference, as well as appropriate sensitivity, were all notable sensor characteristics. Following that, low-cost metal oxide-decorated graphene hybrid materials were created and employed to detect various analytes. Liu et al., for example, used nickel oxide nanoparticle-anchored graphene composite film-modified glassy carbon electrode to detect ascorbic acid selectively [98]. The combination of graphene's excellent electrical conductivity and NiO's outstanding electrocatalytic activity resulted in a sensor with high catalytic activity. The sensor was used to detect the amount of ascorbic acid in vitamin c pills, and the recovery for ascorbic acid quantification ranged from 94.4 to 107.8% for three samples.

#### *Nanocomposites via conducting polymers*

Conducting polymers are copolymerized with nanomaterials along with graphene, carbon, metallic nanoparticles, different beneficial dopants (montmorillonites, clays, and zeolites), and biopolymers (chitosan, chitin, gelatin, cellulose, and proteins) in quite a few

approaches to shape nanocomposites that enhance the combined properties of every issue for higher detection in each electrochemical and biosensing applications. Nanocomposites are an inescapable subset of hybrid substances which might be created through cautiously adjusting the experimental situations and combining more nano-dimensional entities.

These nanocomposites have a substantial impact on improving and rationalizing applications in a variety of scientific and technological domains. Because of their quick responsiveness to changes in current, voltage, and resistance, electrical sensing platforms, such as cyclic voltammetry, amperometry, potentiometry, capacitors, and chemo resistance, and field-effect transistors are used. Due to their overall performance in energy conversion, emission devices, integrated circuit—primarily based totally nano-devices, clinical devices, and different fields, nanomaterial—primarily based totally arrays of sensors are the maximum famous for the manufacturing of electrochemical sensors, biosensors, gas sensors, and immunosensors. Addition of metallic nanoparticles into the host of carrying out polymeric matrix forms accomplishing polymer nanocomposites having advanced electrical, magnetic, thermoelectrically, and biosensing properties. Insertion of metal oxide nanoparticles into the matrix of conducting polymers has been widely studied along with the effects of variation in their dose and dimensions.

The most appropriate technique for enhancing and combining mechanical and electrical properties of carbon nanomaterials (CNMs) such as SWCNT, MWCNT, CNF, fullerenes, carbon nanospheres, graphene (G), and graphene oxide (GO) is embedding them into the polymeric matrix. Carbon nanomaterials can be utilized to manufacture solar cells, nanocomposites materials, electrochemical capacitors, electrochemical sensors, transistors, biosensors, and gas sensors, among other things, as a result of these features. Super capacitor electrodes, light-emitting diodes, transistors, electrochromic devices, electrochemical capacitors, photovoltaic cells, biosensors, gas sensors, actuators, and other uses have all been discovered [99].

Nanocomposites used for electrochemical detection methodology of serotonin

Serotonin, also known as '5-hydroxytryptamine', is a crucial transmitter in the human body that regulates

a variety of central and peripheral activities. 'Serotonin' is likewise a regulator of each energy build-up and intake component of the energy balance. Serotonin regulates hunger and, as a result, nutrient intake at the level of the central nervous system. Electrochemical sensors for serotonin detection are of great interest due to the fact that they provide attractive characteristics consisting of easy sample preparation, less expensive, excessive sensitivity and selectivity, and simplicity of operation [99]. The electrochemical sensors were efficaciously used to discover and quantify serotonin in a lot of physiological fluids, together with entire blood, serum, cerebrospinal fluid, and urine, the usage of most effective a tiny amount of samples and minimum pre-treatment. Electrochemical detection strategies consist of some of the attractive traits that cause them to be beneficial in analytical practice, together with fast evaluation times, cheaper costs, the usage of low-concentration aqueous solutions, and easy sample preparation.

Serotonin levels in body fluids are essential as it is a biomarker for a variety of illnesses, including depression, carcinoid tumors, and diabetes [100]. Normal ranges of serotonin in human fluids are within the nanomolar scale in serum samples, urine, and/or cerebrospinal fluid, and those ranges had been attained utilizing nanomaterials and nanocomposites—primarily based totally sensors. There are kinds of electrochemical sensors: enzymatic sensors known as biosensors and non-enzymatic sensors. The previous is based on enzyme bio-catalytic activity, whereas the second is based on the electrocatalytic qualities of sensitive materials, which favor electro-oxidation responses.

Traditional electrodes, namely gold, glassy carbon, and platinum electrodes, were used to make the first non-enzymatic sensors. Even if these materials are harmless, the large voltage required to electro oxidize serotonin in the measure of the functioning electrodes frequently causes fouling of the detection interface due to oxidation product absorption on the electrode surface. The unfavorable practice alters the working electrode's surface, lowering sensitivity, selectivity, and reproducibility [101].

Significant nanomaterials, e.g., polymeric/metallic composites, noble metals, hydroxides, transitional metals/metallic oxides, and carbon nanocomposites, have been developed, described, and employed to considerably reduce the electro-oxidation potential of serotonin. Voltammetric techniques are commonly

used for the electrochemical discovery of serotonin. The most commonly utilized methods which are CV, SWV, and DPV are shown in the figure. Cyclic voltammetry is a technique for getting information about complex reactions at the electrode surface that is highly beneficial for early electrochemical studies of unknown systems [102].

The voltage between the functioning electrode and a reference electrode is adjusted over interval in the case of SWV, following a pulse pattern. Moreover, the presence of nanostructures in the sensitive layer promotes selectivity, as evidenced by the particular steric selective interaction between serotonin and the active center from the sensor surface, for example. The measurement of serotonin in bodily fluids is extremely useful in medical diagnosis. As a result, it is necessary to develop and correlate traditional approaches with those based on electrochemical sensors.

### Nanocomposites induced CNTs/rGO for serotonin detection

An electrode of glassy carbon (GC) adjusted MWCNTs and Nafion/Ni(OH)<sub>2</sub> is discussed in this article. The electrochemical oxidation of dopamine and serotonin was studied using CV, DPV, and chronoamperometry with a Nafion/Ni(OH)<sub>2</sub>-MWCNTs/GCE sensor. During the recognition of serotonin and dopamine in the existence of ascorbic acid, the reformed electrode acted as an effective electron facilitator. The anodic peaks of dopamine and serotonin may be prominent with the usage of voltammetric techniques, and interference from ascorbic acid changed into a minimum in dopamine and serotonin studies. Upon localization of the anodic height currents, the attention range was found to be 0.05–25 mol•L<sup>-1</sup>, with a detection limit of 0.05 mol•L<sup>-1</sup> for dopamine and 0.008–10 mol•L<sup>-1</sup> with a detection limit of 0.003 mol•L<sup>-1</sup> for serotonin, consistent with data obtained from differential pulse voltage measurements.

The electrode is a treasured sensor in serotonin electroanalysis due to its easy fabrication method, wide linearity range, small detection limit, robust repeatability, and high stability [103, 104]. Another work emphasizes on a new stable and sensitive electrochemical sensor created by combining poly-alizarin red S (AzrS) and MWCNTs with a GCE. Different

electrochemical methods were used to investigate the electrocatalytic oxidation of serotonin in a 0.1 M phosphate buffer solution of pH6 using the chemically modified sensor (poly-AzrS/MWCNTs/GCE). In the detection of serotonin, the electrochemical sensor is effective at mediating electron transport between the electrochemical process and the electrode.

In the case of instantaneous discovery of two analytes in the same solution, the sensor also exhibits properly segregated oxidation peaks for serotonin and adrenalin. The effect of experimental factors on electrochemical behavior was the subject of this research (concurrent detection, the potential scan rate, accumulation interval, pH level in the solution, and concentration). Serotonin has detection and quantification limits of 1.8 10<sup>7</sup> mol•L<sup>-1</sup> and 17.52 10<sup>7</sup> mol•L<sup>-1</sup>, respectively. The electrochemical sensor outperformed the unmodified glassy carbon electrode in terms of sensitivity, stability, and reproducibility in the direct measurement of serotonin in human serum samples [105] as schematically presented in Fig. 11.

Layer-by-layer deposition and electrochemical deposition were used to create the modified GCE. Scanning electronic microscopy was utilized to analyze the morphology of the altered electrode surface, while electrochemical characterizations were performed using cyclic voltammetry and electrochemical impedance spectroscopy. The sensor could be utilized to analyze dopamine and serotonin in biological samples, as well as for medical diagnostics [106].

### Conducting polymer-based electrochemical sensors for uric acid detection

As it is the end product of various metabolic pathways, uric acid levels in the human body give information regarding metabolic abnormalities or disorders such as metabolic syndrome, hypertension, renal injury, and cardiovascular issues. Dai et al. stated that a composite of poly tetra phenyl porphyrin, polypyrrole, and graphene oxide (p-TPP/PPy/GO) coated onto a glassy carbon electrode was employed for uric acid detection, yielding a limit of detection of 1.15 M with a linear range of 5–200 M by DPV in PBS solution at pH 7 [106, 107]. Azimov et al. [108] produced -α-Fe<sub>2</sub>O<sub>3</sub>/polyaniline nanotubes (α-Fe<sub>2</sub>O<sub>3</sub>/PANI NTs) for uric acid sensing by employing poly tetra phenyl porphyrin to increase the electrocatalytic activity of the analytes' oxidation. A 10-h

static synthesis of acetic acid, methanol, aniline, and ammonium persulfate yielded polyaniline nanotubes.  $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$  and polyaniline nanotubes were then stirred together to produce  $\text{Fe}_2\text{O}_3$ /polyaniline nanocomposites. A TEM analysis and FESE determination confirmed the formation of poly tetraphenyl porphyrin—nanotubes accompanied by hexagonal and sphere-shaped- $\text{Fe}_2\text{O}_3$  nanoparticles, which increased electrode surface area. Rajabi et al. reported PBS solution at pH 7; a differential pulse voltammetry sensor was used to determine uric acid concentrations ranging from 0.01 to 5 M, with a limit of detection of 38 nM [109]. An actual urine sample was used to measure uric acid, with recovery values ranging from 98.58 to 101.98%.

#### Conducting polymer-based electrochemical sensors for dopamine detection

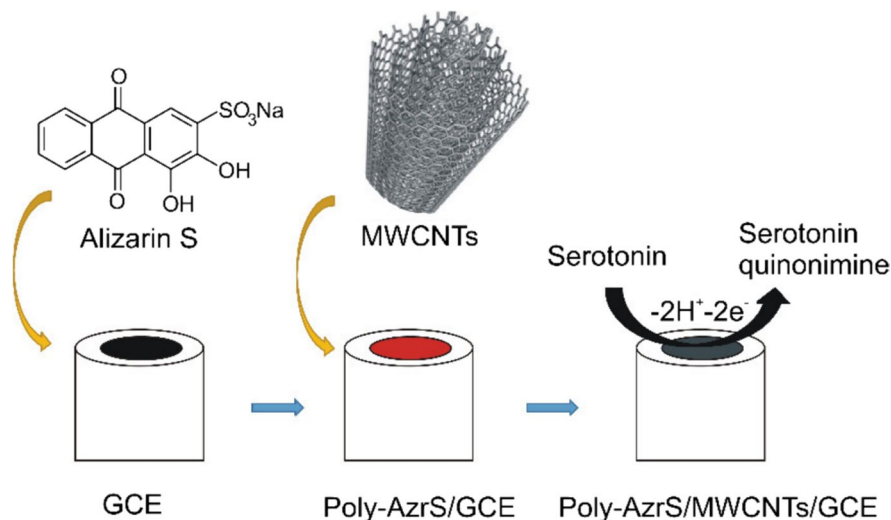
Due to the distinctive physical and chemical characteristics, namely stability, adaptability, versatility, and sensitivity to variations in their electrochemical action as a consequence of minor changes in their surface, intrinsically conducting polymers (CPs) are highly relevant and used for sensor modification. This occurs due to immobilization transfers of the physicochemical characteristics of the modifier to the electrode surface, resulting in huge surface area, tremendous thermal conductivity, and high conduction. By using conducting polymers and carbon nanotubes as modifying materials, electrochemical sensors have advanced. Variety of nanoparticles can also be

appended to polymers to make composites, permitting for the mixture of qualities that can increase the electrical, mechanical, and optical characteristics of the polymer without losing its process ability or addition too much weight.

In addition to their noble film-forming characteristics, electrical conductivity, clear transparency in the visible range, and eco-friendly and thermal stability. Conducting polymers such as polypyrrole, polyaniline, and polythiophene have attracted much attention. Various research clusters with significant efforts in electroanalytical and materials science have developed conducting polymer-based electrochemical sensors to identify of a wide spectrum of analytes [110].

Neurotransmitters are chemicals that transfer neurological impulses and allow neuron cells to communicate with one another. The concentration of these chemicals in the body has an impact on cognitive function, emotion, professional guidance, and physical performance. They are involved in the processes of consciousness, motivation, and memory. It indicates that maintaining human health and preventing mental disorders and diseases requires a proper balance of neurotransmitter concentration in the body. Body biomarkers have emerged as a useful tool for preventing, detecting, and treating a variety of illnesses and degenerative conditions. Because neurotransmitters regulate various actions in cells and tissues, they are one of the most significant biomarkers [111]. Dopamine (DA) is a chemical involved in the regulation of the cardiovascular, renal, and central neurological and hormonal systems. Abnormal

**Fig. 11** Poly-AzrS/multi-walled carbon nanotubes (MWCNTs)/glassy carbon electrode (GCE) sensor for serotonin detection (Copyright © 2021, MDPI) [105]





dopamine levels have been linked to diseases and disorders such as cancer, Parkinson's disease, Huntington's disease, dementia, and drug addiction. Polypyrrole films doped with anionic sulfonated cyclodextrin (PPy-SCD) were potentiostatically deposited on platinum electrodes by Harley, C.C et al. [112].

The resulting films revealed a structure with ridges and valleys that form a ladder-like pattern. At NaCl solutions, a limit of detection of 1 M was obtained chronoamperometrically for dopamine. Furthermore, due to a strong contact between the cyclodextrin dopant and the protonated DA, this modified electrode displayed great selectivity for DA. Chen, X. et al. [113] developed a hybrid sensor based on electrochemically reduced graphene oxide and electropolymerized polypyrrole. In a GO/PBS solution at pH 7.4, cyclic voltammetry will become used to deliver reduced graphene. The potential becomes carried out to the pyrrole approach to deposit polypyrrole. SEM research discovered laminated and spherical structures in pristine PPy/ERGO deposits (attributed to PPy). With the addition of carbonyl groups, a rough, homogeneous, and compact thin film was formed after over-oxidation in a NaOH solution.

Amperometric experiments yielded a value of 0.2 M with a linear response between 0.4 and 517 M, resulting in a value of 0.2 M. The adsorption of positively charged dopamine was increased by a negatively charged sensor surface. For DA detection, ZnO nanotubes supported on molecularly imprinted polymer arrays (MIPs/ZNTs/FTO glass) were used by Wang, H. et al. The Zn nanorods (ZNRs) were deposited onto fluorine-doped tin oxide (FTO) using potentiostat techniques. A low-temperature alkaline solution was then used to chemically etch the zinc oxide nanorods into Zn nanotubes (ZNT). A solution comprising the monomer, lithium perchlorate, and DA was used to electrodeposit polypyrrole sheets. Finally, to oxidize and remove the embedded DA, the electrode was potential dynamically cleaned in PBS. Cylindrical Zn nanotubes (ZNT) covered with PPy sheets were visible in SEM images. The molecular printing approach was found to have a high selectivity for DA.

#### Avenue of carbon nanodiamonds (NDS)

Nanodiamonds (NDs) are carbon nanostructures composed of carbon atoms that are  $sp^3$ -hybridized. They are suitable for certain biosensing applications

owing to their distinctive optical and electrical characteristics. Nanodiamonds provide unique characteristics when compared to carbon nanotubes, such as less toxicity, biocompatibility, refractive index, chemical inertness, and thermal conductivity as well as their very varied surfaces that include functional clusters, namely hydroxyl, carbonyl, ether, and carboxyl groups. Nanodiamonds encompass semi-octahedral structures with faceted surfaces, which may be charged, and chemical functional agencies that may be created through covalent or non-covalent mechanisms on their surface. Nanodiamond surfaces can be created by covalent approaches, but they require a great deal of work. Meanwhile, for the functionalization of ND surfaces, non-covalent changes with an easy process are commonly used [114].

#### Industrial utilization of nanodiamonds for biosensor

Zhang et al. stated that aligned NDs are adequately hydrogenated to conjugate with antibodies via UV-alkene chemistry and that a greater sensitivity to *E. coli* O157:H7 sensing can be achieved at a concentration of 106 cells /mL. The array of three interdigitated electrodes (IDEs) for seeding with NDs was made with 200-nm Au contacts and a 25-nm Cr adhesion layer, with each finger being nine meters thick and nine meters. This means that between the electrode fingers, the ND seeds form electrically conductive islands. The electrochemical detection of DNA based on diamond nanowires (NWs) was pioneered by Yang et al. [115]. Diamond nanoparticles were used as a hard mask to synthesize these metal-like NWs from boron-doped single-crystalline CVD diamond. This group created wires with a length of 3–10 nm and a spacing of 11 nm. Due to anchoring DNA molecules on these wires would yield a density of DNA of around 1012/cm<sup>2</sup> and great effectiveness of DNA sensing, NWs separated by approximately 11 nm were chosen.

The phenyl groups employed to attach the DNA molecules to the diamond were electrochemically functionalized at the nanowire tips. DNA detection on diamond-based biosensors has been investigated using  $[Fe(CN)_6]^{3/4}$  redox mediators. A cyclic voltammogram after exposure to single-base mismatched DNA was analyzed. Because of non-intentional bonding, the amplitude was reduced by around 20% compared to ss DNA. There was,

however, a distinct distinction between complementary ds-DNA and single-base-mismatched DNA bonding. This findings show that  $[\text{Fe}(\text{CN})_6]^{3/4}$  is an excellent predictor of DNA attaching to diamond. Hammers et al. established a label-free biosensor based on covalently bonded DNA oligonucleotides and nanocrystalline diamond sheets with frequency-dependent interfacial electrical characteristics. The DNA molecules changed the impedance of the diamond films by causing a field effect in the diamond space-charge layer [116].

#### Foodborne bacterial detection using cnt's and electrochemical biosensors

Electrochemical detection of food-borne bacterial infections can be augmented by the use of carbon nanomaterial, i.e., CNTs, graphene, and their by-products. Additionally, the carbon nanostructure's electrochemical characteristics enable smooth electron transmission of electrochemical reactions. Biocompatibility and simple connection between the recognition element and target biomolecules have been discovered in carbon nanostructures. Carbon nanomaterial's strong mechanical strength, excellent stability, and simplicity of adjustment make surface functionalization simple [116]. An electronic signal is sent to detect the presence of specific food-borne bacteria and is communicated with biosensor platforms using biorecognition components like DNA, aptamers, antibodies, and enzymes. Biosensor platforms are typically used in conjunction with biorecognition components, namely aptamers, antibodies, DNA, and enzymes to diagnose specific foodborne bacteria and transmit an electronic signal.

The interaction of target analytes with bioreceptors (DNA, antigen, enzymes, antibody, cell structure or cells, biomimetic bases, and bacteriophages) immobilized on an electrochemical transducer delivers quantitative or semi-quantitative analytical information. The electrical signal generated during the interaction between the analytes and the bioreceptor is proportional to the concentration of the analytes. Electrochemical biosensors have a number of advantages over conventional

analytical systems, including the potential to operate with turbid materials, less cost of manufacture, high instrumental sensitivity, and ease of downsizing [117]. In current ages, graphene has been broadly used as a transducer nanomaterial in impedimetric biosensors. Additional advantages of graphene include electron-transfer efficiency, substitute flexible solid substrate, a functionalized surface for quick immobilization of biorecognition proteins, mechanical strength, and metal nanoparticles that have a higher affinity to metal nanoparticles [118, 119].

Electrochemical oxidation in a 0.1 M HCl solution functionalized the graphene screen-printed electrodes with  $-\text{COOH}$  groups. With the  $-\text{COOH}$  group functionalized on graphene surfaces, biorecognition elements can be easily immobilized covalently. The bacteriophages were subsequently easily bound on the graphene surface by forming amide bonds with 1-ethyl-3-(3-dimethyl amino propyl)-carbodiimide (EDC). Non-covalent immobilization of thiolated ssDNA specific to Salmonella outer membrane proteins was used to create a Salmonella aptasensor on a glassy carbon electrode modified with GO and gold nanoparticles [120]. The ssDNA sequence used in this investigation had previously been published. The resistance between the electrode and the electrolyte was used to quantify Salmonella in this investigation. As the bacterial concentration rises, the impedance rises as more bacteria bind to the electrode surface. The detection limit of this detection system was described to be  $3.010 \text{ CFU mL}^{-1}$ , and the lower detection range was attributed to the aptamer's binding specificity and the electron transferability of the GO and gold nanoparticle composite.

Hu et al. advocated collective research of several amplification approaches to improve *E. sakazakii* immunosensing specificity and sensitivity [152]. In their research, they used a graphene and gold nanoparticle (ERGO Au NP) composite to immobilize a horseradish peroxidase (HRP)-labelled antibody specific to *E. sakazakii*. With a detection limit of  $1.2102 \text{ CFU mL}^{-1}$ , these immunosensors were able to detect *E. sakazakii*. The increased sensitivity was attributed to two factors: (1) the presence of HRP, which retained the anti-*E. Sakazakii* antibody's enzymatic activity, and (2) the

electroconductive ERGO Au NP composite, which allowed for effective electron transfer at the electrode–electrolyte interface [121].

Electrochemical biosensors primarily based totally on carbon nanotubes (CNTs) have some attractive features [122]. The graphitic wall of CNT has a greater surface area and surface chemistry, which improves the interactions of different biomolecules and increases the nanomaterial's overall biocompatibility.

Surface functionalization improves the overall solubility and biocompatibility of hydrophobic Bucky tubes of carbon nanotubes. Combinations of various nanomaterials such as metal nanoparticles, polymers, or ionic liquid can be used to functionalize CNT with hydroxyl, carboxyl, or amino groups. Both the basal and edge planes of functionalized CNT show improved solubility, catalytic activity, and surface area. Modified carbon nanotubes with functional groups also allow for the efficient adsorption of biologically recognized molecules (enzymes, DNA, antigen/antibody, and so on) for biosensor applications.

Owing to its special electrical and electrochemical properties, CNT has been identified as promising nanomaterials for use in electrochemical biosensors. CNT-based biosensors are also widely employed due to their inherent benefits, which include high sensitivity, rapid reaction time, and ease of operation, cost-effectiveness, and mobility [123]. In previous studies, MWCNTs have been shown to have a quicker rate of electron transfer, as well as strong redox activity and biocompatibility to a variety of species. To investigate the sensor's interaction with *S. Typhimurium* cells, CV and chronoamperometry were used to characterize it electrochemically. The detection range for this immunosensor was 102 to 107 CFU mL<sup>-1</sup>, with a detection limit of 9.010 CFU mL<sup>-1</sup>.

As a result, tests for inclusivity and exclusivity are required for more reliable detection. CNT can be used in biosensors that are conductometric or impedimetric. This allows for the measurement and monitoring of both electron transfer rate and charges transfer resistance as a result of a biorecognition event. When it comes to detecting *S. Typhimurium*, Dong et al. [144] have proposed a new label-free electrochemical impedance immunosensor based on poly(amidoamine)-MWCNT-chitosan. Gold

nanoparticles were used to immobilize the anti-Salmonella antibody on the electrode. Salmonella cells and antibodies were bound together, allowing for the determination of electron transfer resistance.

With a detection limit of 5102 CFU mL<sup>-1</sup>, this novel combination of nanomaterials improves the sensor's stability, shelf life, and overall sensitivity. When evaluated for interfering effect with other bacteria such as *E. coli* and *S. aureus*, this immunosensor demonstrated good specificity. When the sensor was converted for food analysis, it produced a reasonable result in fat-free milk samples, with a recovery rate ranging from 94.5 percent to 106.6 percent within 1 h of assay time. Additionally, Jia et al. [145] demonstrated that bacterial cells could be detected using reduced graphene oxide (rGO) and carboxyl-modified MWCNTs nanocomposite electrodes. Indeed, electroanalysis having the ability of CNTs further electrochemical reactivity of most essential biomolecules [146–148] and further promote electron transfer reactions of proteins which is most remarkable sensitivity of CNTs to modify the surface conductivity due to the presence of adsorbates permits of significant potential application as highly sensitive nanoscale sensors [151–152].

Various samples are utilized in the form of glassy carbon electrodes as utilized further electro-chemical determination as explored in Table 1. For the detection of Salmonella bacteria, they created a CNT-based impedimetric aptasensor. By using amide bonds, these rGO-MWCNT nanocomposites were covalently bound with amino-modified aptamers specific for Salmonella. The anti-Salmonella aptamers hybridize with their target in the presence of bacterial cells, preventing electron transport and resulting in a bacterial infection.

## Conclusions, outlook, and future aspects

Excellent electrochemical avenues of carbon nanomaterials are widely exploited to detect biomolecules for outstanding biomedical applications. In order to increase the selectivity and sensitivity, the CNMs can be adjusted with a mixture or hybrid or composite of these nanomaterials. It is possible to discuss the many electrochemical characterizations of nanomaterials.

**Table 1** Modified glassy carbon electrodes used for the electrochemical determination of various samples

S. no	Modifier	Method	Linear range	Limit of detection	Samples	Reference
1	ERGO/GCE	DPV	0.5–60 $\mu\text{M}$	0.5 $\mu\text{mol/L}$	Uric acid (UA)	[100]
2	Polystyrene-based resin with graphite powder as new modified carbon paste electrode	DPV,CV,CA	0.3–3.1 $\mu\text{mol/L}$	0.176 $\mu\text{mol/L}$	Urine	[109]
3	PPM-rGO	DPV	0.5–150 $\mu\text{mol/L}$	0.1 $\mu\text{mol/L}$	Urine	[124]
4	PEDOT-modified platinum electrode	CV, LSV	20–100 $\mu\text{mol/L}$	71 $\mu\text{mol/L}$	Urine	[125]
5	SWCNT	DPV	5.0–9.5 $\mu\text{mol/L}$	0.11 $\mu\text{mol/L}$	Tablet and urine	[126]
6	Pt/PANI	CV	0.05–10 $\mu\text{mol/L}$	0.01 $\mu\text{mol/L}$	Uric acid	[127]
7	PGSSG/MWCNT	DPV	1–1200	0.33	Urine	[128]
8	CuNPs/MWCNT-MIP	DPV	0.01–1 $\mu\text{mol/L}$	0.007	Human urine and pharmaceutical	[129]
9	GCE with carbon nanotubes with multiple walls and Nafion/Ni(OH) <sub>2</sub> Nafion/Ni(OH) <sub>2</sub> -MWNTs/GCE	DPV	0.008–10 $\mu\text{mol/L}$	0.003 $\mu\text{mol/L}$	Human blood serum	[130]
10	GCE modified with PEDOT-reduced graphene oxide and silver nanoparticles PEDOTNTs/rGO/Ag NPs/GCE	CV, DPV, CA	1–1.5 $\mu\text{mol/L}$	0.1 $\mu\text{mol/L}$	Bovine assayed multi cera	[131]
12	CNT/PHQ/CE	DPV	0.005–20 $\mu\text{mol/L}$	0.000221	Human blood serum	[132]
13	Nanocomposite material SnO <sub>2</sub> -SnS <sub>2</sub>	CV, SWV, EIS	0.1–700 $\mu\text{mol/L}$	45	Human blood serum	[133]
14	ZnO-Pd/CNTs	DPV	0.6–100 $\mu\text{mol/L}$	0.08 $\mu\text{mol/L}$	Blood serum and human urine	[134]
15	CuO/graphene (CuOG)-modified glassy carbon (GC) electrode	CV	1–8 $\mu\text{mol/L}$	1 $\mu\text{mol/L}$	Glucose	[91]
16	NiO/Ppy	CV	3.0–8.5 $\mu\text{mol/L}$	0.33 $\mu\text{mol/L}$	Glucose	[135]
17	CuO NP – graphene	CV	0.5–2000 $\mu\text{M}$	0.09 $\mu\text{M}$	Glucose	[136]
18	Ni/Cu/MWCNT	CV	25 nM to 800 $\mu\text{M}$	25 nM	Glucose	[137]
19	L-lysine/MWCNT	CV	1 $\mu\text{M}$ to 3.6 mM	8 nM	Hydrogen peroxide	[138]
20	Cobalt oxide/rGO	CV	5 $\mu\text{M}$ to 1 mM	0.2 $\mu\text{M}$	Hydrogen peroxide	[139]
21	Au/PANI	DPV, CV	30–120 $\mu\text{mol/L}$	0.1 $\mu\text{mol/L}$	Dopamine	[140]
22	Cu(II)/AgNP/GCE 78	DPV, CV	2.81 – 8.3 $\mu\text{M}$	0.82 nM	Dopamine	[141]
23	AuNP/PPy	Amperometry	0.1–6.0	0.075	Dopamine	[142]
24	PG/ZnO/CNTs/CPE	DPV	5.0–50.0 $\mu\text{mol/L}$	0.08	Pharmaceutical preparations and urine	[143]

Because of the electrocatalytic activity of CNT-based electrodes, amperometric biosensing of substrates has been possible. In the biomedical field, carbon nanotubes have been utilized to identify biomolecules. Without the need of catalysts, graphene and its derivatives have been utilized to detect biomolecules. For

the detection of serotonin ascorbic acid, dopamine, urine uric acid, DNA, and glucose, the CNTs and graphene electrode performed better. The sensor has demonstrated a lesser limit of detection, high sensitivity, and selectivity to the detection of glucose, H<sub>2</sub>O<sub>2</sub>, DNA, and thrombin.

## Declarations

**Conflict of interest** The authors declare no competing interests.

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