Recent findings in Graphene Modified Electrodes for improved dopamine sensing

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Abstract

Because to its remarkable conductivity and large surface area, graphene and its derivatives have been widely used for electrochemical detection of the neurotransmitter-dopamine. For the detection of dopamine, modified graphene and graphene-based nanocomposites have demonstrated improved catalytic activity. Modification methods include adding heteroatoms and mixing with other nanomaterials, among others. This article summarises and highlights recent advances in graphene-based electrodes for dopamine electrochemical detection. It also seeks to give a general overview of the advantages of employing polymers, metal oxides, and other substances as a linker platform to develop graphene-based nanocomposites for electrochemical dopamine sensors. It also emphasises recent developments of chitosan biopolymer incorporated in graphene nanocomposites and its attractive properties, offering a highly sensitive platform, as well as wearable and implantable DA electrochemical sensors that could be used in diagnosis or the development of therapeutic agents. These developments are based on recent articles published in the last five years.

Keywords

Dopamine, Electrochemical Sensor, Modified Electrodes, Nanocomposites, Polymers.

1. Introduction

Natural molecules called neurotransmitters are essential for the transmission of nerve impulses across synapses from one neuron to another. Dopamine (DA), the most prevalent catecholamine and a key neuromodulator that affects a variety of brain functions and neural development, is one such neurotransmitter. Moreover, it controls vital processes like motion, consciousness, stress reactions, attention span, learning, the sleep-wake cycle, motivation, and memory formation. For a healthy person, the optimum DA content in extracellular fluid should fall between 0.01 and 1 M [1-3]. Any departures from these concentrations may result in physiological and neurological conditions such schizophrenia, HIV infection, addiction, and Parkinson's disease. As a result, accurate and reliable DA detection in physiological fluids is essential for therapeutic implications. Although several approaches have been developed for this purpose, electrochemical techniques have emerged as a cost-effective, highly selective, and fast response approach [4]. Electrochemical techniques are particularly suited for the detection of DA since it is an electroactive compound that is easily oxidizable.

Although being highly sensitive, analytical methods including fluorescence spectrometry, capillary electrophoresis, high-performance liquid chromatography (HPLC), and ultraviolet-visible spectrophotometry demand expensive hardware, trained operators, and a lot of time. Electrochemical procedures have a number of benefits over conventional processes, including simplicity of use, low cost, and the use of inexpensive tools. Several researchers have created electrochemical sensors that exhibit strong affinity for DA, but for practical clinical diagnosis of patients with neurological illnesses, the detection range and limit of detection (LOD) need to be improved. Moreover, the electrochemical DA sensor's selectivity needs to be addressed, especially when working with intricate biological matrices. The accumulation of interferents on the surfaces of unmodified electrodes can cause them to become fouled or poisoned, which can reduce their ability to be reused, selected, and replicated. Ascorbic acid (AA) and uric acid (UA), which have oxidation potentials similar to DA, are the macromolecules in physiological fluids that cause the most interference. By adding the proper materials to the electrode surface, researchers have successfully bypassed these restrictions [5]. Because to its superior conductivity, biocompatibility, and substantial specific surface area, graphene and its derivatives, notably carbon-based nanomaterials, have been extensively employed in the development of DA sensors. Graphene-containing nanocomposites have demonstrated excellent performance in the electrochemical analysis of DA. The biomedical applications of graphene and its composites for dopamine detection are described in the figure 1 and 3 and its evolution for measuring dopamine in figure 2. Graphene-based nanocomposite materials have been demonstrated to connect well with chitosan, a non-toxic natural polymer with lots of hydroxyl and amine groups, developing a novel nanocomposite with good dispersion properties, improved catalytic activity, and biocompatibility for usage in actual media [6–9].

Recent research has demonstrated the effectiveness of graphene and its derivatives in detecting DA in the presence of AA and UA. When compared to other carbon-based nanomaterials, graphene offers superior physical and electrical properties. Graphene is a single-layered carbon atomic sheet that is structured in a hexagonal lattice configuration. It is the perfect choice for developing electrode sensing materials for a variety of analytes due to its stability, high surface area to volume ratio, and potential window [10–11]. The most recent findings on graphene and modified graphene electrodes for the detection of DA are summarised in this study.

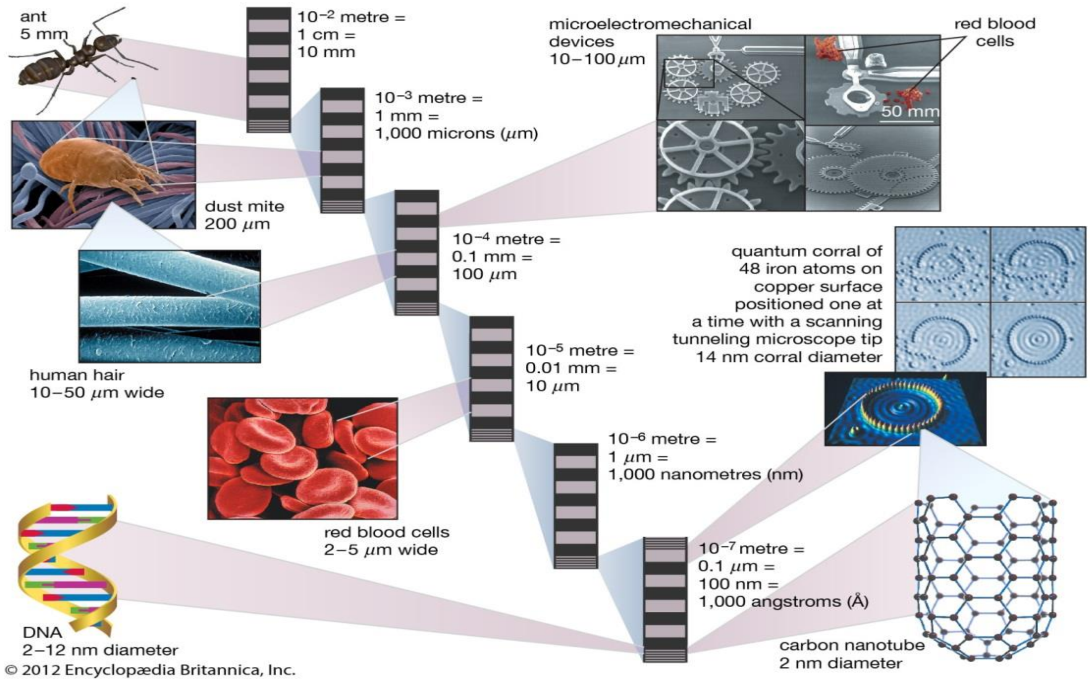

Fig.1. Biomedical Applications of Graphene

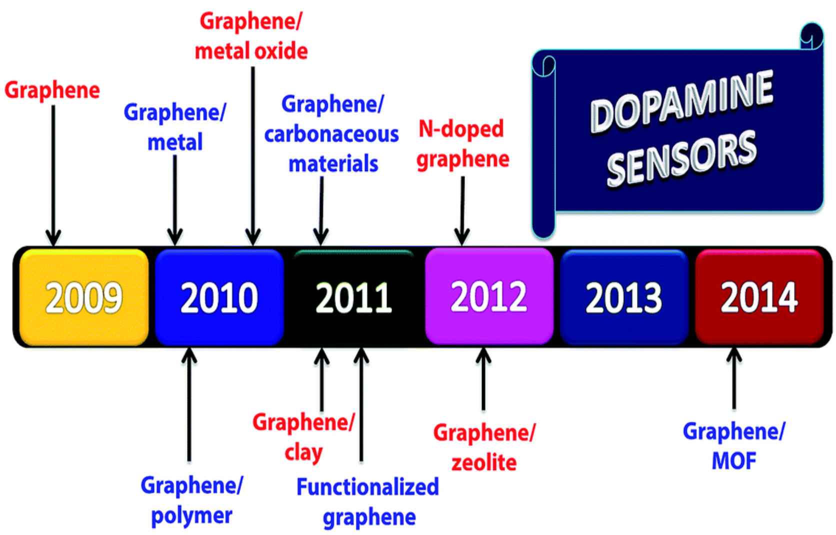

Figure 2 Evolution of Dopamine sensors

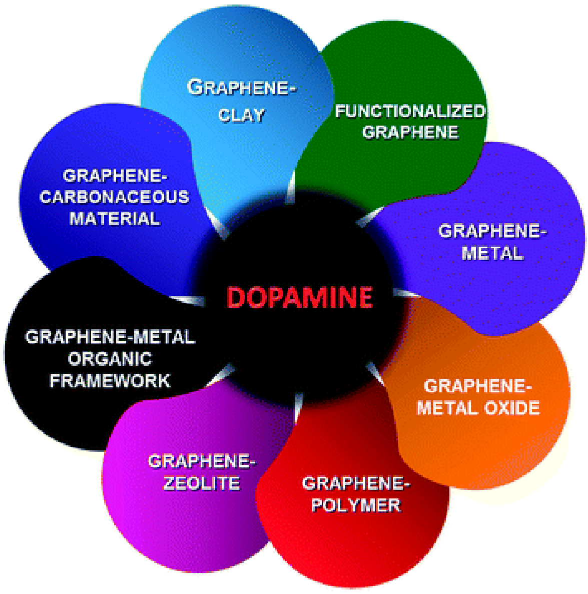

Figure 3 Graphene and its composites materials in dopamine detection [12]

2. Application of Graphene in Electroanalytical Detection of DA

2.1 Reduced Graphene Oxide

Reduced graphene oxide (rGO), one of the graphene family's members, is regularly utilized as an electrode material for electrochemical sensors. It comes from graphene oxide (GO), which is an oxygenated form of graphene with a high concentration of functional groups that contain oxygen, such as -OH, -COOH, and epoxy groups. However, the poor conductivity of GO limits its application in electrochemical sensors. When GO is partially reduced to rGO, its conductivity is boosted and its conductive carbon conjugated networks are restored. After reduction, rGO still contains a small number of oxygenated groups that allow for dispersion in aqueous solutions and make it easier for electrons to transfer during electrochemical detection [12–14]. This capability also allows for in-place composite formation. While reduced graphene of the type produced by Hummer's approach is frequently employed for the electroanalytical detection of DA, other varieties have also been produced and used as modified electrodes [15]. Using electrochemically reduced graphene nanoribbons, for instance, one example is the simultaneous and individual electrochemical assessment of DA and UA (ERGNRs). When multi-walled carbon nanotubes were longitudinally oxidatively unzipped, graphene oxide nanoribbons (GONRs) were produced. These GONRs were then electrochemically changed to produce ERGNRs [16]. The resulting SPCE/ERGNRs sensor displayed selective simultaneous detection of DA and UA with LOD of 0.15 M (S/N = 3) and 0.3 M (S/N = 3), respectively, and achieved good peak to peak separations (DA-UA = 245 mV).

This study demonstrates the feasibility of designing simple, scalable graphene-based sensors with high sensitivity and specificity for the detection of DA in complex sample matrices. The ability to detect DA at such low concentrations has considerable implications for clinical diagnosis and patient monitoring of persons with neurological diseases, because DA levels can be predictive of disease progression or treatment response. More investigation is required to improve the sensor's performance and design, as well as to look into the feasibility of adapting this method to detect other analytes. The versatility and potential of graphene-based sensors for a range of electrochemical sensing and biosensing applications are highlighted in this work.

2.2 Doped Reduced Graphene Oxide

Due to problems including graphene sheet aggregation and uneven dispersion, electrodes treated with reduced graphene oxide (RGO) frequently have poor electrochemical characteristics. Researchers have been experimenting with a variety of solutions to this issue, such as adding defects, functional groups, or doping chemicals to improve the conductivity and electrochemical responsiveness of graphene. Due to nitrogen's resemblance in size to carbon and its capacity to form bonds with carbon atoms thanks to its five valence electrons, nitrogen doping has become a common procedure. As a result, N-doped reduced graphene oxide (N-rGO), which performs noticeably better than undoped rGOs, has emerged as the preferred material for electrochemical detection of dopamine (DA) was found from recent studies. Researchers have experimented with a variety of synthesis strategies, including chemical and hydrothermal approaches, to create N-rGO nanoparticles for modified electrodes in DA sensing [19–20]. For use in dopamine (DA) electrochemical sensing, several methods for synthesising nitrogen-doped reduced graphene oxide (N-rGO) have been devised and investigated. Soni et al., describes a low-temperature, economically advantageous photocatalytic method for the synthesis of N-rGO. In this procedure, aqueous GO and ammonia solution are combined as a N dopant precursor, stirred, and then exposed to UV light for 60 minutes to facilitate the photo-catalytic reduction of RGO. High temperatures, which might lead to the aggregation of graphene sheets, are not necessary for this process. In comparison to rGO modified GCE, the resulting N-rGO coated on glassy carbon electrodes had a larger linear range and lower limit of detection, as well as a strong activity towards DA oxidation. In order to determine ascorbic acid (AA), DA, and uric acid (UA) electrochemically, Feng et al. produced a holey nitrogen-doped graphene aerogel (HNGA) [21–24]. To achieve the ideal pyridinic N content at 800°C, the HNGA was synthesised using a hydrothermal process in the presence of H_2O_2, lyophilized, and annealed in a mixture of ammonia and argon gases. Excellent electrochemical responses to AA, DA, and UA were seen in HNGA-modified electrodes, and the three-dimensional porous structure of HNGA is what explains the quick heterogeneous electron-transfer kinetics.

2.3 Graphene Nanocomposite

The contact between the functionalized molecule and the graphene single layer makes it simple to functionalize graphene by chemical bonding or physical adsorption. Due to this property, graphene nanocomposites can be made, which have extraordinary physicochemical qualities that surpass those of their constituent parts. Current studies show that due to homogeneous dispersion and material intercalation between the graphene layers, graphene nanohybrids spread more steadily than graphene alone. Graphene nanocomposites improve electroanalytical characteristics and sensing capability by increasing graphene dispersibility. Since they enhance electron transfer kinetics and electrocatalytic activity for the oxidation of DA, metallic graphene nanocomposites and polymeric graphene nanocomposites have drawn increasing interest from scientists. Many examples of graphene/metal nanoparticle and graphene/polymer nanocomposites for electrochemical detection of DA are found in the literature [28].

2.4 Graphene/Metal Oxide

Subramaniam et al. synthesized a nanocomposite with reduced graphene oxide (rGO) enclosed in delafossite using a simple wet-chemical technique (CuAlO2). Due to the interaction energy between the CuAlO2 nanostructures and graphene sheets, the nanocomposite demonstrated improved electrochemical performance when compared to a pure CuAlO2 electrode in the detection of DA. A key element in the nanocomposite's high performance is the regulated activity of its porous structure. By avoiding interference from UA and AA, the suggested rGO/CuAlO2/GCE electrode demonstrated outstanding performance, with a nanomolar range limit of detection, good stability, and reproducibility [29]. Wang et al. developed a magnetic graphene nanocomposite (Fe2O3-NiO@GO) for the electrochemical detection of DA using the electrodeposition method. With a linear range of 10 to 1500 M, a detection limit of 0.005 M, and a sensitivity of 0.16812 A/M, the Fe2O3-NiO@GO/GCE platform showed good affinity for the oxidation of DA. Due to the catalytic properties of the carbon nanostructures and magnetic nanoparticles contained in the nanocomposite, the nanocomposite also demonstrated a wider linear range of DA determination and equivalent detection limit values to existing graphene-based electrochemical sensors. The practical effectiveness of Fe2O3-NiO@GO/GCE in detecting DA in actual samples of human blood serum was also examined by Wang et al. [30]. It has been shown that a nanocomposite of nano-octahedral-shaped Mn3O4 and reduced graphene oxide (rGO/Mn3O4) exhibits enhanced electrocatalytic activity towards the simultaneous detection of DA and UA. The nanocomposite with a molar ratio of 1/10 shown improved electrocatalytic activity, leading to outstanding catalytic activity towards the simultaneous detection of DA and UA with a broad linear range and the lowest LOD. The rGO/Mn3O4/GCE electrode that was suggested demonstrated high stability, acceptable repeatability, good selectivity, and the capacity to detect DA and UA in biological residues and pharmaceutical samples [25–30].

2.5 Graphene/Metal Nanoparticles

As a platform for the simultaneous detection of DA, AA, and UA, Minta et al. developed a nanocomposite made of N-doped reduced graphene oxide/gold nanoparticles (N-rGO/AuNPs). GCE/N-rGO-Au, the modified electrode, was made by electrodepositing AuNPs on its surface. With well-resolved anodic peaks and a good peak-to-peak separation, the modified electrode showed adequate electrocatalytic oxidation activity towards the three analytes. The homogeneous and well-distributed AuNPs were electrodeposited onto the N-doping RGO due to the great affinity of the metallic particles to the nitrogenated regions of the graphene material. The electrical conductivity, catalytic activity, and sensing properties towards AA, DA, and UA were all increased by the uniform distribution of AuNPs [25–30].

3. Graphene/Conducting Polymers

Researchers have created composite materials using conducting polymers and graphene that have improved electroanalytical activity for dopamine (DA). In one study, Hsine and colleagues developed nanohybrids with redox conduction and anionic functionalities that inhibit non-specific contacts and overlapping using chemically reduced graphene oxide (CRGO) and modified poly(para-phenylene) (Fc-ac-PPP). These nanocomposites showed a well-resolved redox peak oxidation and could detect AA, DA, and UA both separately and together. The sensors were reliable and reusable for identifying these substances in urine samples, offering a strong diagnostic platform. In a different work, Teng et al. used hybrids of conducting polymer poly(3,4-ethylenedioxythiophene) (PEDOT) and nitrogen-doped graphene (NG) for the electrochemical detection of DA. Due to the combined features of nitrogen doping, which prevents graphene sheet stacking, and PEDOT, which increases the hybrids' conductivity while reducing NG stacking, these nanohybrids shown improved electrocatalytic activity towards DA oxidation. With a low detection limit of 54 nM and a detection range from 0.2 M to 90 M, the synergistic interaction between NG and PEDOT created a rapid electron transfer rate with electrocatalytic activity towards DA oxidation. These nanohybrids continued to be very stable and selective towards DA while displaying distinct redox signals for DA, AA, and UA.

4. Graphene/Chitosan Nanocomposite

There are many benefits of using chitosan in graphene-based nanocomposites for electrochemical sensing. Chitosan is a natural biopolymer that is non-toxic, biocompatible, and biodegradable, making it suitable for use in biomedical applications. Chitosan has large amounts of hydroxyl and amine groups, which can be easily modified by conductive polymers, graphene sheets, and nanoparticles to produce composites with enhanced electron transfer rates and targeted surface areas [33]. Furthermore, under the right circumstances, the amino groups in chitosan can form strong imine or amide linkages with carbonyl groups, which greatly increases the stability and repeatability of the electrochemical sensor. The chitosan can become cationic when amino groups are present, it is simpler for the desired biomolecule to interact electrostatically with the chitosan matrix. Finally, because of its hydrophilic properties and functional groups, which improve charge transfer ability, chitosan is a desirable material for electrochemical sensing. Nanocomposites, which have higher dispersion and superior chemical properties, are made by combining chitosan and graphene. The resulting flexible membranes can be used in flexible patches or biostrip sensors. When utilized in graphene-based nanocomposites for electrochemical sensing, chitosan has a variety of advantages that can improve the analytical performance of the sensor [30-35].

Koyun et al. modified a glassy carbon electrode with a nanocomposite made of reduced graphene oxide, chitosan, and chromium oxide in order to construct a sensor. The modified electrode with well-separated voltammetry peaks allowed for the simultaneous detection of dopamine, uric acid, xanthine, and hypoxanthine. A platform consisting of chitosan and graphene sheets was developed by Begum et al. for the simultaneous detection of dopamine and uric acid. The scientists chemically pre-treated the graphene sheet to enhance the amount of carboxylic acids on its surface in order to maximize the degree of amidation between chitosan and graphene. The resulting chitosan-graphene sheet platform demonstrated better electrocatalytic activity due to a higher amount of amide functionalization, which resulted in preference determination towards dopamine and uric acid in numerous real samples. In many aspects, chitosan's chemical properties make it a good material for making sensors, however even when mixed with graphene, it lacks conductivity and electroactivity. In order to boost sensitivity, current sensor research focuses on integrating graphene with other materials, where each material provides unique properties to form efficient electrodes for electroanalytical sensors. Composite graphene and chitosan have been combined with different polymers and nanomaterials to improve conductivity and electron transport capabilities [30–35].

5. Conclusion

The development of effective electrochemical sensors for detecting DA in real-world applications depends on the use of materials with particular electrical and structural properties. Graphene, a material with these characteristics, has an atomically thin sheet-like structure, a large surface area, the ability to transmit electrons, is biocompatible, and has a preference for biomolecules. Recent improvements in production methods, heteroatom doping, and graphene incorporation with other nanomaterials have improved its electrical and electrochemical capabilities. The focus of this review is on high-performance electrochemical sensors that use graphene and its cutting-edge derivatives and nanocomposites as interfaces to detect DA. It also discusses recent advancements in the chitosan biopolymer, which is a key component of graphene nanocomposites and offers a highly sensitive platform for developing wearable and implantable DA electrochemical sensors that may be utilised for therapeutic or diagnostic treatments.

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