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*Review Article*

# Graphene Oxide Modified Electrodes for Dopamine Sensing

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Dopamine (DA) is one of the most important catecholamine neurotransmitters that plays an important role in the central nervous, renal, hormonal, and cardiovascular systems. Since its discovery, tremendous effort has been made and various techniques have been developed for the DA detection. Recently, graphene-based materials have attracted a tremendous amount of attention due to their high sensitivity and rapid response towards effective detection of DA. This review focuses on current advances of graphene-based materials for DA detection based on recent articles published in the last five years.

## 1. Introduction

Dopamine (DA) is an important catecholamine neurotransmitter in the mammalian central nervous system and plays a very important role in memory, hormonal, and cardiovascular systems [1–4]. The imbalance or low levels of their contents are likely to cause neurological disorder such as senile dementia, HIV infection, schizophrenia, and Parkinson's disease [5–10]. Therefore, the accurate and rapid determination of DA at submicromolar concentration level is crucial for diagnoses and monitoring and gained immense attention among numerous researchers. In the last few decades, tremendous effort has been made on developing analytical methods with a high sensitivity and good selectivity for detecting DA in biological samples.

In physiological processes of human metabolism, ascorbic acid (AA), dopamine (DA), and uric acid (UA) are seen as crucial small biomolecules and well known to coexist in biological matrixes such as blood and urine as well as in extracellular fluid at a high concentration level. Moreover, due to almost similar potential of DA, AA, and UA, they can be oxidized easily and cause great interferences on the bare working electrode resulting in poor response resolution in DA determination. Thereby, simultaneous determination of DA, AA, and UA by electrochemical method is necessary and have been reported widely.

Due to the similarity of oxidized potentials of DA, uric acid (UA), and ascorbic acid (AA), many electrochemical methods have been established for simultaneous determination in their mixture [11–19]. However, these analytical techniques have some limitations, such as being time-consuming, low-sensitivity, complicating pretreatment, and being of high in cost. Recently, chemical modified electrodes have been developed and reported to effectively detect DA with enhanced sensitivity and selectivity [20–23]. Among them, graphene is extensively used to fabricate electrochemical sensors and hold a great promise as ideal candidate sensing platforms. Graphene is one of the most promising and versatile materials discovered ever, of which the first truly two-dimensional material is its single sheet that shows many outstanding properties including high charge mobility, high thermal and electrical conduction, transparency, and good mechanical properties [24–28]. Considering the characteristic of the graphene and its excellent properties, graphene-based nanomaterials [29, 30], chemically reduced graphene oxide (CR-GO) [31], sulfonated graphene (s-GR) [32], chitosan-graphene [33], and electrochemically reduced graphene oxide (ERGO) [34], have been reported to effectively detect DA, UA, and AA.

In this review, attention is focused on summarizing and highlighting the categories of graphene-based materials and their application for the detection of dopamine.

## 2. Electrochemistry of Graphene

The basic electrochemical behaviors, like electron transfer rate, electrochemical potential window, redox behavior, and

Table 1: Comparison of some characteristics of the different modified electrodes for the determination of DA.

| Electrode | Linear range ($\mu$M) | Detection limit ($\mu$M) | Reference |
|---|---|---|---|
| Ag-Pt/nanofiber | 10–500 | 0.11 | [55] |
| MWCNT/CCE | 0.5–100 | 0.31 | [56] |
| HNCMS/GCE | 3–75 | 0.02 | [57] |
| PtNPs-MWCNT/GCE | 0.06–2.03 | 0.05 | [58] |
| OMC/Nafion | 1–90 | 0.5 | [59] |
| PDDA@HCNTs/GCE | 2.5–10 | 0.08 | [23] |
| Nanostructured gold | 10–100 | 5 | [60] |
| 2-Amino-thiazol film/GCE | 5–25 | 5 | [61] |
| Meso-$SiO_2$/CPE | 0.4–25 | 0.1 | [62] |
| CAT/ZnONps/CPE | 5–41 | 3 | [63] |

MWCNT/CCE: multiwalled carbon nanotube modified carbon-ceramic electrode; HNCMS/GCE: hollow nitrogen-doped carbon microspheres modified glassy carbon electrode; PtNPs-MWCNT/GCE: Pt nanoparticles decorated multiwall carbon nanotubes modified glassy carbon electrode; OMC: ordered mesoporous carbon; PDDA@HCNTs: poly(diallyldimethylammonium chloride) functionalized helical carbon nanotubes; Meso-$SiO_2$/CPE: mesoporous silica nanoparticles/carbon paste electrode; CAT/ZnONps/CPE; catalase ZnO nanoparticles modified carbon paste electrode.

so forth, are important parameters and should be studied first for any potential application. The high electron mobility at room temperature [35] and a low resistivity at low temperatures [36] are the excellent electronic qualities of graphene and have been a much studied subject. The electrochemical properties of graphene, such as well-defined redox peaks [37] and low charge resistance [38], are also of high contemporary interest.

Graphene-based electrodes exhibit well-defined redox peaks, of which the redox processes are predominantly diffusion-controlled [37, 39, 40]. The unique electronic structure of graphene with high density of the electronic states and the surface physicochemistry of graphene are beneficial for fast electron transfer [37, 41, 42]. A variety of oxygenated species possessed by the edges of graphene sheets can significantly increase the rate of electron transfer by creating specific surface functional groups [43]. The edge-plane defect sites on graphene with high density provide multiple active sites for electron transfer to biospecies [44]. Compared to graphite, graphene has significantly more uniform distribution of electrochemically active sites and its special 2D structure making it very efficient in detecting adsorbed molecules [45, 46].

## 3. Electrochemical Detection of Dopamine

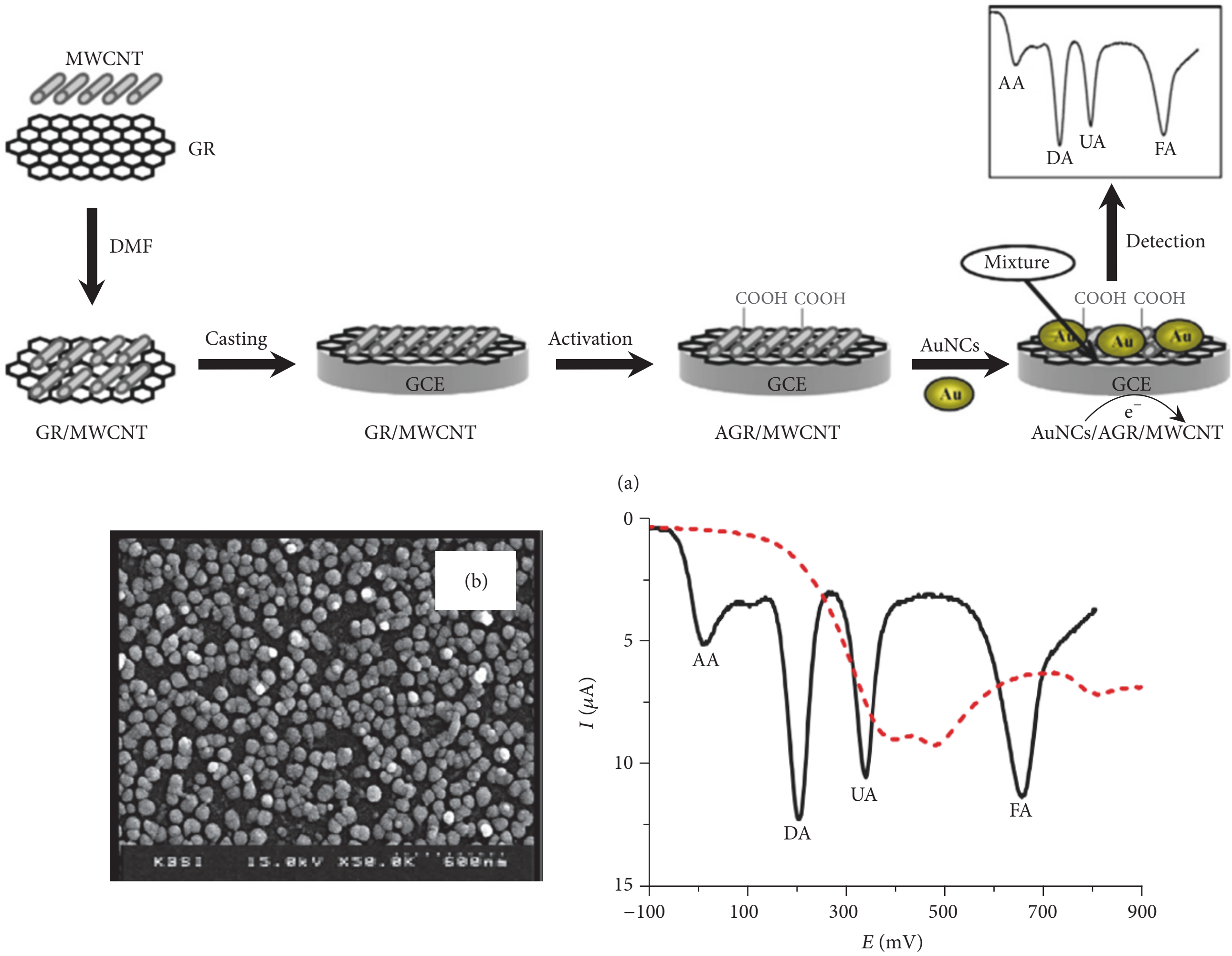
It turned out that catecholamines are very convenient models for studying the mechanism of the metal-molecule interaction and determining the adsorption of these flat molecules on single crystal metal surfaces. Dopamine (DA) is one of the most important catecholamine neurotransmitters in the mammalian central nervous system and a topic of much interest in the development of electrochemical sensors [47–49].

Due to its electroactivity, DA molecules can be easily oxidized on the electrode. In the last few decades, DA detection has gained much attention. Rapid, simple, and sensitive electrochemical methods with modified electrodes have shown promising platform for detection of DA. Numerous materials, such as metal nanoparticles, carbon nanotubes, fullerenes, polymers, graphene, and enzymes, have been used as modifiers to construct highly sensitive and selective DA biosensors to distinguish the coexistence of DA, AA, and UA in a biological environment [50–54]. Table 1 shows analytical data of DA detection in presence of AA and UA obtained from use of different modified electrodes.

## 4. Graphene Oxide (GO) Modified Electrode for Dopamine Sensing

Graphene, a "rising-star" carbon material has attracted enormous interest in science communities and a promising candidate in many fields such as nanomaterials, supercapacitors, nanoelectronics, catalysis, nanophotonics, and sensors. Owing to its unique electronic, mechanical, and thermal properties, graphene has been intensively employed as modification materials on the surface of different electrodes. Up to now, many modified electrodes supported by graphene oxide, such as graphene-metal oxide nanocomposite, GO-carbon dot composite, GO-bimetallic nanoclusters, layer-by-layer assembled multilayer films of GO/metal nanoparticles, GO/polymer nanosheets, and GO/hybrid polymer composite [64–74], have been successfully prepared for the electrochemical detection of dopamine and have shown their unique properties. Even so, the development of novel graphene combined with AuNPs nanocomposite for the electrochemical determination of dopamine remains interesting and challenging.

*4.1. Nanostructured Material Modified GO.* An electrochemical sensor using activated graphene (AGR)/MWCNT nanocomposite loaded Au nanoclusters (AuNCs) was developed by Abdelwahab and Shim for the simultaneous determination of DA, AA, UA, and folic acid (FA) [75]. AuNCs modified electrode was reported to provide a stable large surface area that effectively improved sensitivity of the sensor. The details fabrication of AuNCs/AGR/MWCNT nanocomposite sensor was presented in Figure 1(a). SEM image shows the growth and attachment of AuNCs onto the AGR/MWCNT layer with an average diameter of 50 nm (Figure 1(b)). The proposed nanocomposite sensor showed satisfactory results




(b) (c)

Figure 1: Schematic representation of the fabrication of AuNCs/AGR/MWCNT nanocomposite sensor (a). SEM image showing the particle size of the fabricated AuNCs/AGR/MWCNT/GC (b). SWV of bare (dashed line) and AuNCs/AGR/MWCNT (solid line) electrodes in 0.1 M PBS (pH 7.0) containing 30 $\mu$M AA, 40 $\mu$M DA, 20 $\mu$M UA, and 30 $\mu$M FA (c) [75].

and sharp oxidation peaks of AA, DA, UA, and FA as compared to a bare electrode during SWV measurement as shown in Figure 1(c).

On the other hand, Hu et al. have used reduced graphene oxide- (rGO-) carbon dots (CDs) composite film for sensitive detection of DA with concentration in the range from 0.01000 uM to 450.0 uM with the detection limit as 1.5 nM [76]. An excellent environment for DA oxidation was reposted using the rGO-CDs composite that also acts as a barrier to prevent other interfering substances during DA detection. The CDs had carboxyl groups with negative charge, which could make the surface of the electrode more conductive and enabled interaction and electron communication between rGO and DA. Other researcher developed a novel gold nanoparticles/tryptophan-functionalized graphene composite (AuNPs/Trp-GR) modified electrode that exhibited excellent electrocatalytic properties towards the oxidation of dopamine [66].

One-step electrodeposition of Pt–Au bimetallic nanoclusters loaded onto graphene oxide- (GO-) electrochemically reduced GO (ERGO) graphene (Au–Pt/GO–ERGO) was developed and investigated by Liu at al. for sensitive and simultaneous detection of DA and UA [77]. It was observed that Au–Pt/GO–ERGO provide better mass transport of reactants to the electrocatalyst compared with other metal anchored GO. The modified electrode showed a good anti-interference ability, reproducibility, and high stability. The electrochemical performance of the carbon nitride nanosheets-graphene oxide (CNNS-GO) composite was investigated for simultaneously detection of AA, DA, and UA by Zhang et al. [78]. A remarkable electrocatalytic activity was observed with their composite modified electrode towards determination of DA with low detection limit (0.096 uM) and good sensitivity. Figure 2(a) shows the possible response mechanism of graphitic CNNS-GO composite. It was demonstrated that, after modification of

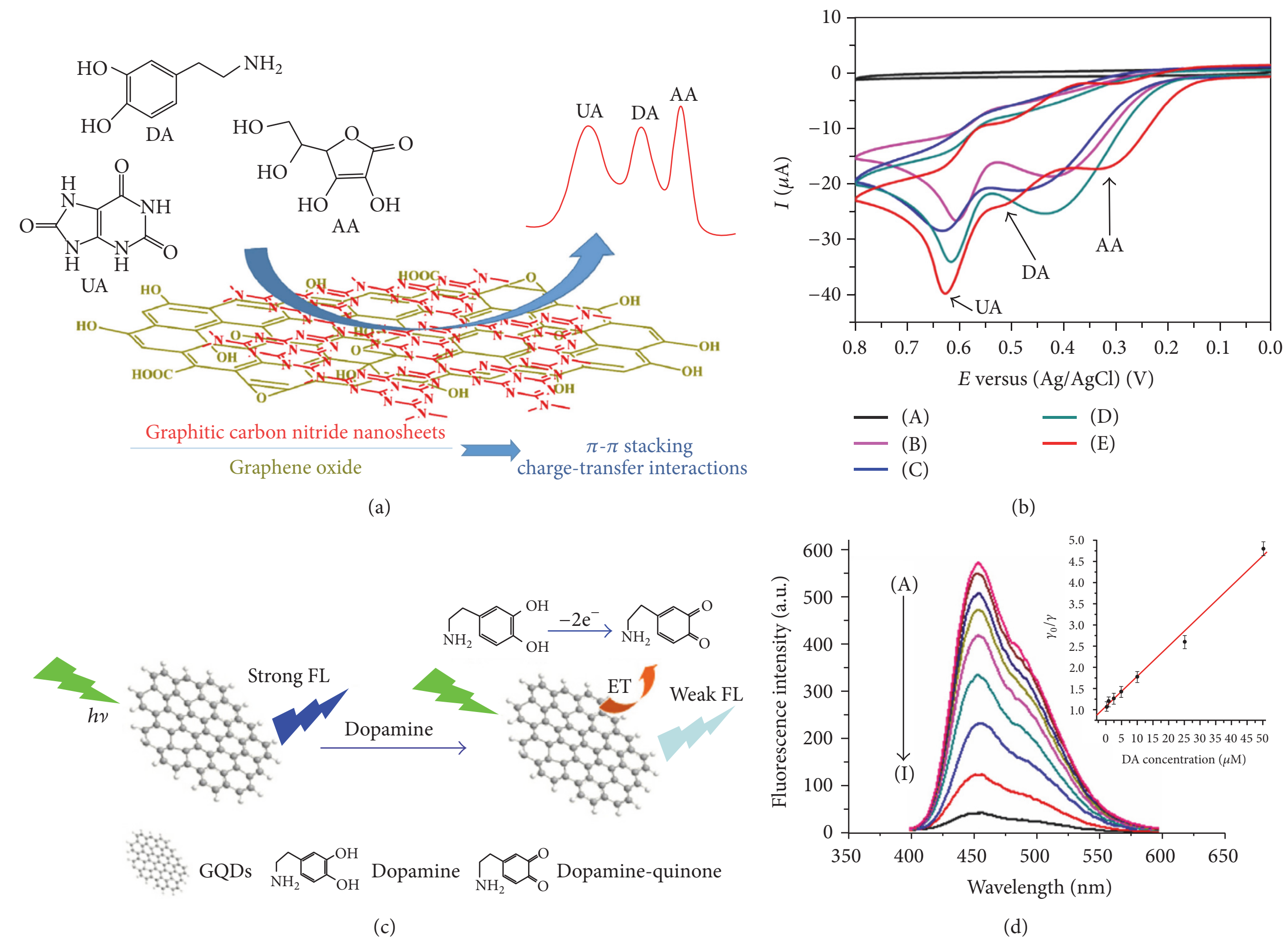



Figure 2: (a) Schematic drawing of electrochemical oxidized AA, DA, and UA on CNNS-GO/GCE. (b) CV of 0.01 M PBS with pH 4.0 on CNNS-GO/GCE (A), CV of 2 mM AA, 0.1 mM DA, and 0.5 mM UA in 0.01 M PBS with pH 4.0 on bare GCE (B), CNNS/GCE (C), GO/GCE (D), and CNNS-GO/GCE (E) [78]. (c) Schematic illustration of the developed graphene quantum dots based fluorescence sensing system for DA detection [79]. (d) Fluorescence emission spectra of the sensing system after addition of various concentrations of DA ((A)–(I): 0, 0.25, 1, 2.5, 5, 10, 25, 50, and 100 $\mu$M). Inset: the relationship between the florescence quenching and the concentration of DA (0.25, 1, 2.5, 5, 10, 25, and 50 $\mu$M).

GCE with CNNS-GO, the peak potential of AA, DA, and UA clearly separated as shown in Figure 2(b).

A promising dopamine sensor based on $Fe_3O_4$@graphene nanospheres (GNs) modified glassy carbon electrode was developed by Zhang et al. [73]. It was proved that the surface of $Fe_3O_4$@GNs/Nafion/GCE can easily attract the positively charged aromatic DA molecules through $\pi$-$\pi$ stacking and electrostatic attraction. In addition, a simple and suitable method was explained for the quantitative determination of micromole level of DA. Zhao et al. have developed a label-free fluorescence-based method using graphene quantum dots (GQDs) for selective and sensitive detection of dopamine (DA) [79]. Considering excellent biocompatibility, GQDs are superior in comparison with conventional semiconductor QDs. Figure 2(c) represents the possible mechanism of fluorescent sensing strategy for the detection of DA. The fluorescence spectra of the GQDs in the presence of different concentrations of DA were shown in Figure 2(d).

Another promising electrochemical sensor based on novel graphene-tantalum wire (Gr/Ta) electrodes was prepared by Zhao et al. with electrochemical deposition of MgO nanobelts (denoted as MgO/Gr/Ta) for the simultaneous detection of AA, DA, and UA in biological fluid [80]. The strong electrocatalytic properties of MgO nanobelts with large surface area can increase the peak separation between analytes during simultaneous determination of AA, DA, and UA (Tables 2 and 3).

*4.2. Polymer Modified GO.* Recently the design of novel nanocomposites with combination of graphene and conductive polymers plays a challenging role in elaboration of hybrid electrodes with (bio)sensing properties. Due to long-term environmental stability, strong adherence to electrode surface, high electric conductivity, large active sites, good selectivity, and biocompatibility, polymer modified electrodes have been paid increasing attention and become a research focus [65, 83, 84].

Table 2: Comparison of some characteristics of the different nanostructured modified GO electrodes for the determination of DA.

| Electrode | Linear range ($\mu$M) | Detection limit ($\mu$M) | Reference |
|---|---|---|---|
| AuNCs/AGR/MWCNT | 1–210 | 0.08 | [75] |
| rGO-CDs/GCE | 0.01–450 | 0.015 | [76] |
| AuNPs/Trp-GR | 0.5–411 | 0.056 | [66] |
| Au–Pt/GO–ERGO | 0.06–498 | 0.02 | [77] |
| $Fe_3O_4$/r-GO/GC | 0.4–3.5 | 0.08 | [81] |
| Au/RGO/GCE | 6.8–41 | 1.4 | [17, 18] |
| GQDs–$TiO_2$ | 0.02–105 | 0.067 | [82] |
| CNNS-GO | 1–20 | 0.096 | [78] |
| $Fe_3O_4$@GNs/Nafion/GC | 0.02–130 | 0.007 | [73] |
| MgO/Gr/Ta | 0.1–7 | 0.15 | [80] |

AuNCs/AGR/MWCNT: gold nanoclusters/activated graphene/MWCNT nanocomposites; rGO-CDs/GCE: reduced graphene oxide-carbon dot composite; AuNPs/Trp-GR: gold nanoparticles/tryptophan-functionalized graphene nanocomposites; Au–Pt/GO–ERGO: Au–Pt bimetallic nanoclusters decorated on graphene oxide-electrochemically reduced GO; $Fe_3O_4$/r-GO/GC: $Fe_3O_4$ magnetic nanoparticles/reduced graphene oxide nanosheets modified glassy carbon; Au/RGO/GCE- Au nanoplates and reduced graphene oxide modified glassy carbon electrode; GQDs-$TiO_2$: graphene quantum dots-$TiO_2$ nanocomposites; CNNS-GO: carbon nitride nanosheets doped graphene oxide; $Fe_3O_4$@GNs/Nafion/GC: Nafion covered core-shell structured $Fe_3O_4$@graphene nanospheres modified glassy carbon electrode; MgO/Gr/Ta: MgO nanobelt-modified graphene-tantalum wire electrode.

Table 3: Comparison of some characteristics of the different nanostructured modified GO electrodes for the determination of DA.

| Electrode | Linear range ($\mu$M) | Detection limit ($\mu$M) | Reference |
|---|---|---|---|
| AuNP/Gr/OPPy-MIP/GCE | 0.5–8 | 0.1 | [65] |
| Si-MG-MIP | 8–200 | 1.5 | [83] |
| GO-Ag/PLL/GCE | 0.1–10 | 0.03 | [84] |
| PILs/PPy/GO | 4–18 | 0.07 | [67] |
| PEDOT/RGO | 0.1–175 | 0.03 | [86] |
| PAM/rGO/GCE | 0.3–50 | 0.1 | [87] |
| PFeTPP/PSS-Gr | 0.1–40 | 0.05 | [72] |
| PPy/GQDs | 0.005–8 | 0.0001 | [74] |

AuNP/Gr/OPPy-MIP/GCE: gold nanoparticles, graphene, molecularly imprinted overoxidized polypyrrole; Si-MG-MIP: silanized magnetic graphene oxide-molecularly imprinted polymer; GO-Ag/PLL/GCE: graphene oxide-Ag/poly-L-lysine modified glassy carbon electrode; PILs/PPy/GO: poly(ionic liquids) functionalized polypyrrole/graphene oxide nanosheets; PEDOT/RGO: conducting polymer poly(3,4-ethylenedioxythiophene) doped with reduced graphene oxide; PAM/rGO/GCE: polyacrylamide/reduced graphene oxide nanocomposites; PFeTPP/PSS-Gr: poly[iron (II) tetraphenylporphyrin] and poly(sodium-p-styre-nesulfonate) modified graphene; PPy/GQDs: polypyrrole/graphene quantum dots core hybrids.

Do et al. have developed a biosensor for rapid detection of DA with graphene (Gr) films and molecularly imprinted (MIP) overoxidized polypyrrole (OPPy) modified electrode [65]. The prepared electrode showed a good recognition capacity for template molecule (DA) in the presence of other structurally similar molecules (AA, UA). The preparation of AuNP/Gr/OPPy-MIP/GCE electrodes, involving four main steps, is shown in Figure 3(a). The CV (cyclic voltammograms) and DPV (differential pulse voltammograms) responses of electrochemical signals to different DA concentrations in the presence of 1 mM AA and 50 $\mu$M UA are demonstrated in Figures 3(b) and 3(c). A reduced graphene oxide- (RGO-) polypyrrole composite electrode was developed by Fritea et al. for sensing of catechol and dopamine [85]. They reported that their developed electrode could have promising applications in the miniaturization of amperometric biosensors for in vivo measurements of low level of neurotransmitters. Similarly, Guo et al. have developed graphene oxide-Ag/poly-L-lysine modified glassy carbon electrode (GO-Ag/PLL/GCE) for the determination of DA in the presence of AA. The GO-Ag/PLL/GCE electrode improved the DA electrochemical catalytic oxidation, which demonstrates that GO-Ag/PLL/GCE has a remarkable electrocatalytic activity towards the oxidation of DA [84]. Figure 3(d) represents the immobilization and hybridization of DA on the GO-Ag/PLL. The excellent electrocatalytic activity of GO-Ag/PLL/GCE towards the oxidation of DA is also investigated by DPVs as shown in Figure 3(e). It was reported that, under the optimum condition, the modified electrode showed good selectivity, excellent linear relation from 0.1 to 10 $\mu$M for DA, and high sensitivity with a detection limit of 0.03 $\mu$M for DA from their mixture solution.

On the other hand, recently a novel composite consisting of poly[iron (II) tetraphenylporphyrin] (PFeTPP) and poly(sodium-p-styre-nesulfonate) (PSS) modified graphene (Gr), denoted as PFeTPP/PSS-Gr, was developed by Zhang et al. [72]. It was demonstrated that the fabricated composite exhibits superior electrocatalytic activity towards the oxidation of DA. Schematic representation of the procedure for the preparation of PFeTPP/PSS-Gr is shown in Figure 4(a).

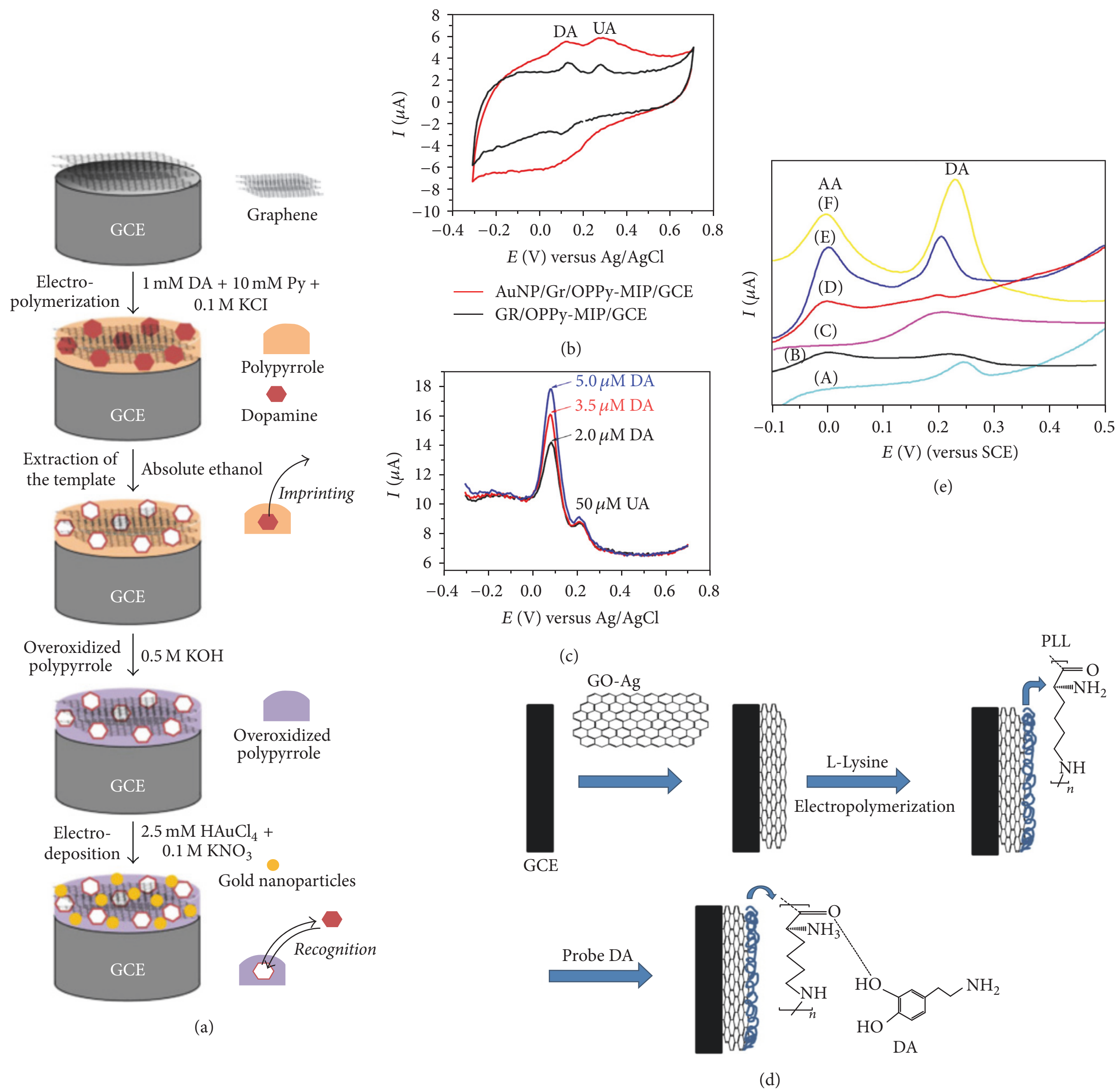



FIGURE 3: (a) Schematic fabrication process of AuNP/Gr/OPPy-MIP/GCE and its recognition for DA. (b) CVs for 1 mM AA, 50 $\mu$M UA, and 10 $\mu$M DA, in 0.1 MPBS at AuNP/Gr/OPPy/GCE. (c) DPVs of mixture solution of different concentrations (2.0, 3.5, and 5.0 $\mu$M) of DA, 1 mM AA, and 50 $\mu$M UA in 0.1 M PBS at AuNP/Gr/OPPy-MIP/GCE. (d) Schematic illustration of the fabrication process of electrochemical sensors for the determination of DA. (e) Differential pulse voltammograms (DPVs) for AA and DA at bare GCE (A), GO/GCE (B), PLL/GCE (C), GO-PLL/GCE (D), GO-Ag/GCE (E), and GO-Ag/PLL/GCE (F).

Clear lattice fringes as shown by HRTEM image in Figure 4(b) reveal good crystallinity of the PFeTPP nanocrystals. They also investigated the electrochemical behavior of a mixture containing 500 lM AA, 30 lM UA, and DA with different concentrations at the PFeTPP/PSS-Gr modified electrode by DPV, and the results are presented in Figure 4(c). It was revealed that the favorable electrostatic attraction and $\pi$-$\pi$ stacking between positively charged DA and negatively charged PFeTPP/PSS-Gr can enhance the interactions of DA molecules to the electrode surface and accelerate the electron transfer [72].

In another work, a sensitive and selective chemiluminescence sensor for dopamine measurement was developed by Duan et al. with silanized magnetic molecularly imprinted polymer (Si-MG-MIP) that was prepared by encapsulating inorganic magnetic particle with organic polymer [83]. The Si-MG-MIP exhibited excellent merits and had shown high selectivity and sensitivity for on-site

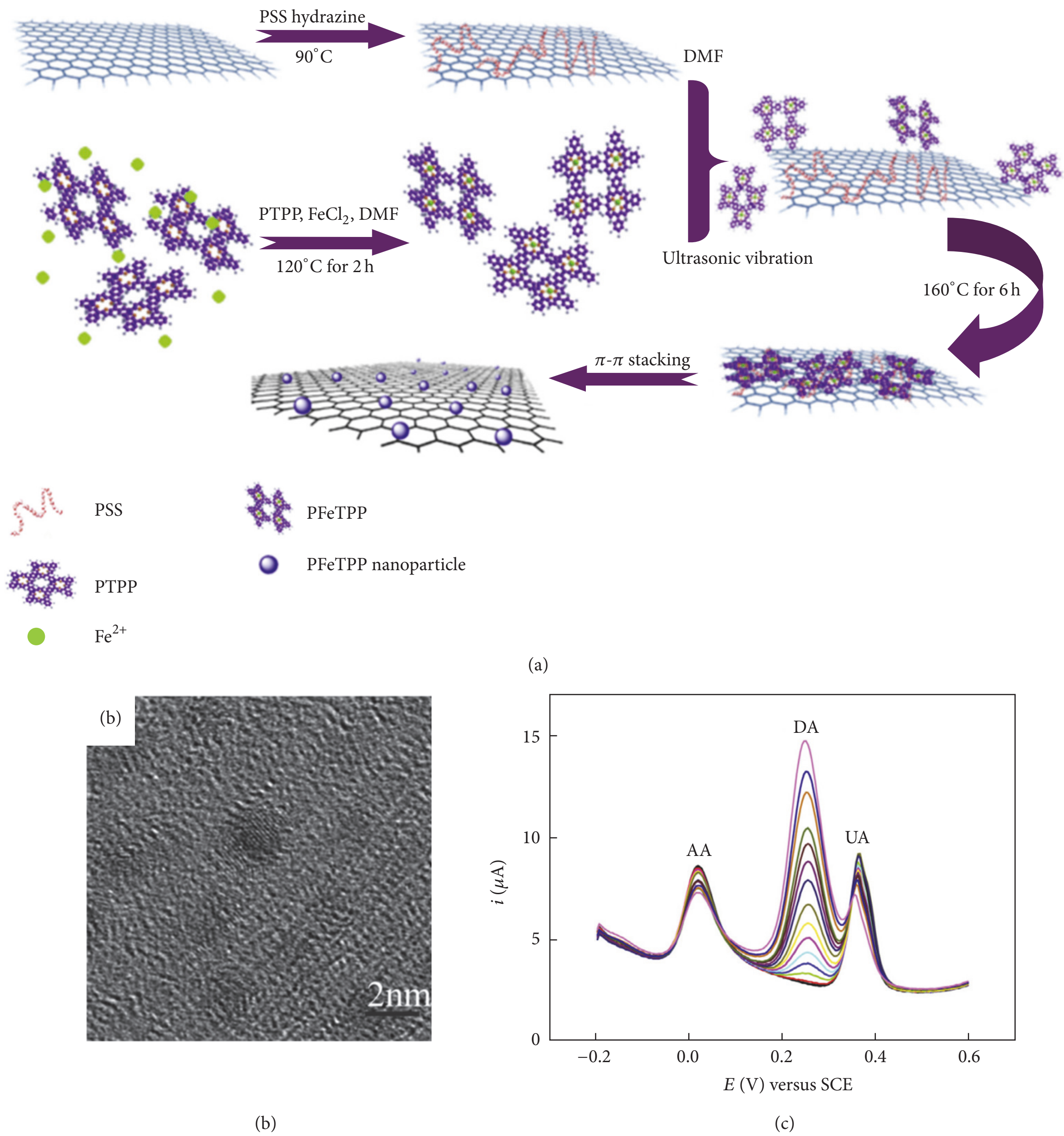
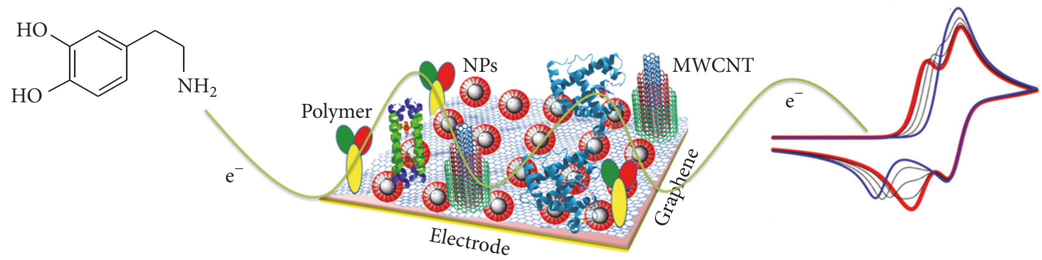



Figure 4: Synthesis routes of the PFeTPP/PSS-Gr composite by the solvothermal method (a). HRTEM images of PFeTPP/PSS-Gr (b). DPVs of DA at the PFeTPP/PSS-Gr modified GCE in the presence of 500 $\mu$M AA and 30 $\mu$M UA with different concentrations (c).

measurement of DA. Novel poly(ionic liquids) functionalized polypyrrole/graphene oxide nanosheets (PILs/PPy/GO) were prepared on GCE, which was reported to show excellent electrochemical catalytic activity, good steady, and sensitivity towards DA sensing [67]. It was revealed that the existence of PILs effectively improved the transmission mode of electrons and resulted in the different electrocatalytic performance towards the oxidation of DA and AA. Wang et al. have developed an electrochemical sensor capable of sensitive and selective detection of DA using a nanocomposite composed of conducting polymer poly (3,4-ethylenedioxythiophene) (PEDOT) doped with GO [86]. Their prepared GO doped PEDOT (PEDOT/GO) nanocomposite, which later on was reduced electrochemically, was reported to detect DA in a wide linear range from 0.1 to 175 $\mu$M, with a detection limit of 39 nM.

## 5. Concluding Remarks

The use of graphene modified electrode in electrochemical biosensors faces explosive growth owing to their sensitive surfaces state and other many excellent characteristics of




Scheme 1: Working principle of graphene-based dopamine sensor. The illustration explains the electron transfer mechanism and sensitivity between DA and monolayer modified semiconductor electrode.

graphene. In this review, the entire outline of recent advances in dopamine sensing by suing graphene-based electrochemical biosensors is presented and discussed (Scheme 1). The great significance of sensitive and selective simultaneous detection of dopamine and its interference electroactive species eliminates the need for new and miniaturized device fabrication. To improve the hybrids electronic and thermal conductivity, the combination of metallic composite materials with GO can be a promising candidate. The recent investigations suggest that graphene oxide-metal nanocomposites were superior in terms of peak current density, linear range, and limit of detection for dopamine sensors. On the other hand, it was observed that GO-polymer modified electrode act as a good steady and sensitive material that effectively improved the transmission mode of electrons and resulted in the different electrocatalytic performance towards the oxidation of DA.

Another area that deserves expanded attention is the interaction of graphene oxide with biological samples and little to the live cells. More biological systems and related ambient conditions of using graphene in different sensing applications need to be addressed. Above all, the graphene-based modified electrode could be an extremely promising candidate applicable for a wide range of biosensing applications. However, further studies of sensing mechanism along with rich information on intermolecular interaction will accelerate development of the advanced neurotransmitter sensors in the near future.

## Conflicts of Interest

The author declares no conflicts of interest.

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