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Different Electrochemical Sensors for Determination of Dopamine as Neurotransmitter in Mixed and Clinical Samples: A Review

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Dopamine (DA) is one of the most important catecholamine neurotransmitters in the human central nervous system of the brain playing a key role in the functioning of the renal, hormonal, and cardiovascular systems. Abnormal levels of dopamine are related to neurological disorders, such as schizophrenia and Parkinson's disease. Accordingly, the control and fluctuations of the amount of dopamine are extremely important in monitoring the analytical systems in the human brain. This review covers the different electrochemical sensors for the determination of dopamine as neurotransmitter and points out the advantages and disadvantages of them.

Keywords: Dopamine, Modified electrodes, Electrochemical detection, Neurotransmitters

Abbreviations: DA, Dopamine; CV, Cyclic voltammetry; CHA, Chronoamperometry; SWV, Square wave voltammetry; EIS, Electrochemical impedance spectroscopy; DPV, Differential pulse voltammetry; AC, Acetaminophen; XN, Xanthine; AA, Ascorbic acid; UA, Uric acid; FA, Folic acid; TRP, Tryptophan; SWCNT, Single walled carbon nanotube; MWCNT, Multi walled carbon nanotube; SPE, Screen printed electrode; GCE, Glassy carbon electrode; CPE, Carbon paste electrode; ITO, Indium tin oxide; GPE, Graphite paste electrode; CF, Carbon fiber; PBS, Phosphate buffer solution; GR, Graphene; RGO, Reduced graphene oxide; CNTs, Carbon nanotubes; QDs, Quantum dots; CoL, Complex N,N'-bis(3-ethoxysalicylidene)-4,5-dimethyl-1,2-phenylenediamino cobalt(II) chloride; CoClSal, 5-Chloro salicylidene) 2,2-dimethyl-1,3-propandiamino cobalt(II) chloride; EDC.HCl, (N-3-Dimethylaminopropyl)-N'-ethylcarbodiimide hydrochloride; HOBt, N-Hydroxy benzotriazole; BzTC, Benzethonium chloride; MoS₂, Molybdenum disulfide; WO₃NPs, Tungsten oxide nanoparticles; 3DGH-AuNPs, Three-dimensional graphene hydrogel and gold nanoparticles nanocomposite; TiN, Titanium nitride; AuNPs, Gold nanoparticles; CuO, Cupric oxide; Co₃O₄, Cobalt oxide; SDS, Sodium dodecyl sulphate; GLN, Gelatin; NiHCF, Nickel hexacyanoferrate; PDAN, Poly 1,5-diaminonaphthalene; PTh, Poly thiophene; BAMB, 1,4-bis (aminomethyl) benzene; Co(OH)₂, Cobalt hydroxide; O, Graphene oxide; GA, Gallic acid

INTRODUCTION

Dopamine (DA) with IUPAC nomenclature 4-(2-aminoethyl)-benzene-1,2-diol, is one of the most important catecholamine neurotransmitters in the mammalian central nervous system in the brain from where dopamine functions as a neurotransmitter that is a chemical substance released

by the nerve cells to send signals to other nerve cells, thus acting as a chemical messenger playing a key role in the functioning of the renal, hormonal, and cardiovascular systems. Abnormal levels of DA are related to neurological disorders, such as schizophrenia and Parkinson's disease [1-7]. In Parkinson's disease, an age-related degenerative condition causing tremor and motor impairment, the dopamine-transmitting neurons in the substantia nigra area die, causing a loss of dopamine-secreting neurons in this

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area. As an intravenous medication dopamine influences on the sympathetic nervous system to produce effects such as increased heart rate and blood pressure. Dopamine, however, cannot cross the blood-brain barrier; therefore when dopamine is given as a drug it does not directly affect the central nervous system. Therefore, L-Dopa, that is the precursor of dopamine and can cross the blood-brain barrier easily, is given to patients with diseases such as Parkinson's disease to increase the amount of dopamine in their brains [8-13]. For diagnosis of these diseases, very accurate measurements of DA in biological samples are required. Electrochemical techniques are most widely used for detection of DA. Low cost of the electrochemical instrumentation advocates a big advantage, however, a perception of low sensitivity and selectivity while dealing with real samples is somehow justifiable. With the development of new materials, researchers have tried chemically modified electrodes for detection of DA. Very low detection limits have been achieved with modified electrodes while such detections were not possible with bare electrodes [14-17]. The present review has sought to explore the technological advances made in recent years towards the selective analysis of DA and critically evaluate how they could, in fact, be exploited as a basis for a multi analyte sensor incorporating DA detection. Numerous strategies have been evolved in recent years mostly focused on the manufacture and response characterization of discrete sensors. Various methods of obtaining selective detection such as use of nanocomposites, carbon nanotubes, graphene and polymers are also discussed along with the clinical relevance of DA determination.

Dopamine Determination by Modified Electrode with Chemical Component

The chemical modification of inert substrate electrodes with mediators offers significant advantages in the design and development of electrochemical sensors. In operations, the redox active sites shuttle electrons between a solution of the analyte and the substrate electrodes often along with a significant reduction of the activation overpotential. A further advantage of chemically modified electrodes is that they are less prone to surface fouling and oxide formation compared to inert substrate electrodes. Esfandiari Baghbanidi *et al.* reported a synthesized 2-(4-oxo-3-phenyl-

3,4-dihydroquinazoliny)-N'-phenyl-hydrazine carbothioamide and used it to construct a modified graphene oxide nano sheets paste electrode. The electrocatalytic oxidation of dopamine at the surface of this electrode was studied using cyclic voltammetry (CV), chronoamperometry (CHA), and square wave voltammetry (SWV). Under the optimized conditions, the square wave voltammetric peak current of dopamine increased linearly with dopamine concentration in the range of 5.0×10^{-7} - 4.0×10^{-4} M. The detection limit of 9.5×10^{-8} M was obtained for dopamine. Finally, this modified electrode was used for determination of dopamine in some real samples [18].

A chemically modified carbon paste electrode with 2,2'-[3,6-dioxa-1,8-octanediy]bis(nitriloethylidene)]-bishydroquinone (DOH) was employed to study the electrocatalytic oxidation of DA in buffer solution using CV, SWV and CHA reported by Mazloum Ardakani *et al.* The diffusion coefficient ($D = 7.4 \times 10^{-6}$ cm² s⁻¹) and the kinetic parameters such as the electron transfer coefficient ($\alpha = 0.33$) of DA oxidation at the surface of 2,2'-[3,6-dioxa-1,8-octanediy]bis(nitriloethylidene)]-bis-hydroquinone modified carbon paste electrode were determined using the electrochemical approaches. It has been found that under an optimum condition (pH 7.0), the oxidation of DA at the surface of the modified electrode occurs at a potential about 290 mV less positive than that of an unmodified carbon paste electrode. The catalytic oxidation peak currents showed a linear dependence on the DA concentration and linear analytical curves were obtained in the range of 3.0×10^{-5} - 2×10^{-3} M of DA with SWV. The detection limit was determined to be 3.2×10^{-6} M. This method was also used for the determination of DA in a pharmaceutical preparation (injection) by the standard addition method [19].

Shahbakhsh and coworkers studied a modified graphite paste electrode with [Sr(OH)₂·4(phen-dione)₂](Cl)₂ complex (Sr-Phen) and used it for simultaneous electrochemical determination of DA, acetaminophen (AC), and xanthine (XN). The proposed modified electrode showed a sensitive and selective response for simultaneous determination of DA, AC and XN to three well separated peaks using DPV method in a phosphate buffer solution with pH 3.0. Some kinetic and thermodynamic parameters for the electrochemical oxidation of DA were also determined. Under the optimum conditions the calibration curve was

linear is 5-220 μM with a theoretical detection limit ($S/N = 3$) of 0.8 μM for DA [20].

Sodium dodecyl sulfate surfactant modified graphene paste electrode, prepared by electrochemical immobilization of the Sodium dodecyl sulfate surfactant on a graphene paste electrode, was applied for simultaneous determination of DA in the existence of ascorbic acid (AA) and uric acid (UA) through CV and differential voltammetry (DPV) suggested by Manjunatha. The modified electrode showed strong electrocatalytic function for the oxidation of DA, AA and UA and three well-defined voltammetric peaks of about 167 mV, 12 mV and 303 mV by CV method. A linear response in the range of 1×10^{-5} - 1×10^{-3} M with a detection limit of 4.7×10^{-6} M and limit of quantification of 15×10^{-6} M for DA was obtained. The effects of graphene paste constitution, concentration, surfactant, pH and scan rate were investigated. The modified electrode was highly stable and could be used to determine DA in injection samples [21].

The electro-oxidation of DA, UA and their mixture on a gold electrode modified by a self-assembled monolayer of 2-(3,4-dihydroxyphenyl)-1,3-dithialone has been studied by Mazloum Ardakani *et al.* The mediated oxidation of DA at the modified electrode under the optimum condition (pH = 7.0) in CV occurs at a potential about 220 mV less positive than that of the unmodified gold electrode. DPV exhibited a linear dynamic range over the concentration range of 0.2-250.0 μM and a detection limit of 0.07 μM for DA. The modified electrode was used for simultaneous determination of DA and UA using DPV. The results showed that the electrode is highly efficient for the catalytic electrooxidation of DA and UA, leading to a remarkable peak resolution (~ 350 mV) for two compounds [22].

Nagles *et al.* synthesized $[(\text{Co}(\text{bdmpzm})_2(\text{NCS})_2)]$ (bdmpzm = bis(3,5-dimethylpyrazol-1-yl)methane) and combined it with single walled carbon nanotubes (SWCNT) and nafion on a screen printed carbon electrode (SPCE). The electrocatalytic activity of modified electrode was evaluated for DA oxidation. Anodic and cathodic peaks, observed at 0.42 V and 0.29 V, respectively, showed a much more quasi-reversible process ($\Delta E = 0.128$ V) compared to the electrode without cobalt complex. The detection limit ($3\sigma/b$) for DA was 0.095 μM . The studies of reproducibility determined as relative standard deviation (RSD) was 2.5%

($n = 5$) with four different electrodes. The repeatability was 1.5% ($n = 5$). The sensor was used in quantifying DA in human urine with an average recoveries between 83.0 and 93.0% [23].

Another research by Uzun and Hastemir for determination of DA was performed using a glassy carbon electrode (GCE) modified with N-(1-H-indole-3yl)methylene thiazole-2-amine (IMT2A). IMT2A was deposited on the GCE by CV. This modified electrode demonstrated an electrocatalytic effect on the oxidation of DA in the presence of UA and AA using DPV method in 0.1 M phosphate buffer solution (PBS) of pH 7. Selective determination was realized in elimination of AA response on the IMT2A modified electrode. The oxidation peak currents increased linearly with two concentration intervals of DA at pH 7 phosphate buffer. One of them is 0.25-9.15 μM , and the other is 9.15-95.1 μM . The limit of detection was found to be 0.086 μM . The proposed electrode was applied to the determination of DA in pharmaceutical preparations and human urine sample with satisfactory results [24].

Gnahor *et al.* described using carbon paste electrode modified with sulfated β -cyclodextrin for determination of DA. The electrode showed high affinity towards DA electrochemical oxidation. DA returned a concentration detection range 5×10^{-7} - 5×10^{-4} M and a detection limit of 1.33×10^{-7} M [25].

Saktehivel *et al.* reported hydrothermal synthesis of molybdenum disulphide (MoS_2) flakes. The synthesized material effectively utilized for the fabrication DA electrochemical sensor. Moreover, the successful formation of flake-like MoS_2 was confirmed by X-ray diffraction (XRD), scanning electron microscope (SEM) and energy dispersive X-ray (EDX) studies. An electrochemical sensor, the flake-like MoS_2 modified GCE, exhibited higher electrocatalytic activity in the oxidation of DA in terms of higher oxidation peak current and lower oxidation potential when compared with bare GCE. The flake-like MoS_2 based electrochemical sensor has been fabricated to detect DA in a wide linear response range (0.006-181 μM), good sensitivity ($3.98 \mu\text{A} \mu\text{M}^{-1} \text{cm}^{-2}$) and very low detection limit of 2 nM. Moreover, the flake-like MoS_2 modified GCE showed a good selectivity even in the presence of biologically co-interfering compounds and common metal

ions [26].

Dopamine Determination by Modified Electrode with Polymer

Conducting electroactive polymers are novel materials which are successfully used or are expected for the use in various fields of technology like batteries, anticorrosion coatings, processing of electronic circuit boards, *etc.* Among many applications of conducting polymers, electroanalysis of solution species seems to be very promising. Electrodes, modified with conducting or electrogenerated polymers, possess many interesting features that can be exploited for numerous electroanalytical and sensor applications.

In a research conducted by Ozcan *et al.*, preparation of a single-use electrochemical sensor for the selective and sensitive determination of DA was investigated by electrochemical polymerization of pyrrole-3-carboxylic acid on electrochemically over-oxidized pencil graphite electrode (p(P3CA)/EOPGE). CV measurements of $\text{Fe}(\text{CN})_6^{4/3-}$ indicated that the electrochemically over-oxidized PGE (EOPGE) showed superior electron transfer characteristics according to bare PGE. The ionized carboxyl groups found in the structure of poly (pyrrole-3-carboxylic acid) (p (P3CA)) showed high affinity towards positively charged DA. The combination of the advantages of EOPGE and p (P3CA) in p (P3CA)/EOPGE led to a synergistic effect on the electrochemical oxidation of DA. The p (P3CA)/EOPGE showed high selectivity towards DA by discriminating its oxidation potential from the common interfering substances such as AA and UA. The p (P3CA)/EOPGE showed linear responses in the electrochemical oxidation of DA between the concentration values of 0.025 μM and 7.5 μM . Detection limit was determined as 0.0025 μM according to signal to noise ratio (S/N:3). Analytical application of p (P3CA)/EOPGE was successfully tested in the determination of DA in blood serum and pharmaceutical samples [27].

Liu *et al.* described the preparation of a molecularly imprinted polymer (MIP) film on the surface of electrodeposited hollow nickel nanospheres (hNiNS), and the use of this in an electrochemical sensor for DA (Fig. 1). The electropolymerization condition such as pH value, functional monomer and ratio of template to monomer were optimized. By using DA as a model analyte, the sensor, if

operated at 0.1 V vs. SCE, has fairly low detection limit of 1.7×10^{-14} M, two wide assay ranges of 5×10^{-14} - 1×10^{-12} M and 1×10^{-12} - 5×10^{-11} M, and superb selectivity [28].

Koyun *et al.* investigated the use of an overoxidized nanofiber polypyrrole (OONfPPy) modified pencil graphite electrode (PGE) as a sensor for the determination of DA. The performance of the modified electrode was studied using DPV method. The calculated electroactive areas of the bare electrode and $\text{OO}_{10}\text{NfPPy}_5\text{PGE}$ were found to be 4.54×10^{-8} cm^2 and 1.05×10^{-6} cm^2 . The sensor ($\text{OO}_{10}\text{NfPPy}_5\text{PGE}$) showed a high selectivity to DA with a detection limit of 6.95×10^{-9} M (S/N = 3). To demonstrate the validity of the sensor for the determination of DA, pharmaceutical and human serum samples were performed [29].

An electrochemical sensor was developed by Kannan and Sevvell for selective and simultaneous determination of DA and paracetamol (PA) using a GCE with electropolymerized poly-4-amino-6-hydroxy-2-mercaptopyrimidine (AHMP) thin film (Poly-AHMP/GCE). The modified electrode showed well-defined redox peaks for DA and PA with enhanced peak currents and reduced over potential while bare GCE showed a broad peak. Poly-AHMP/GCE separated overlapped and sluggish signals of DA and PA into two well-distinguished redox peaks with large enough anodic peaks separation about 240 mV. Under the optimal experimental condition, the anodic peak current of DA increased linearly within the concentration ranges 2.5-25 μM with correlation coefficient of 0.999 and the detection limit was found to be 0.20 μM , (S/N = 3) by DPV method [30].

A conducting polymeric film of trypan blue (TB) was deposited onto an anodized glassy carbon electrode (AGCE) surface by electropolymerization reported by Li *et al.* The poly(trypan blue)-modified AGCE was used for the determination of DA in the presence of UA and AA. The PTB/AGCE exhibited good electrocatalytic behavior for the oxidation of DA in phosphate buffer solution (PBS, pH 7.0). The electrochemical oxidation signals of DA, UA, and AA were well-resolved into the three distinct peaks in cyclic voltammograms with the anodic peak potential separations (ΔE_{p_a}) of *ca.* 186, 145 and 331 mV between AA-DA, DA-UA, and AA-UA. A detection limit of *ca.* 0.36 μM (S/N = 3) was obtained for the sensing of DA with a linear range of 1-

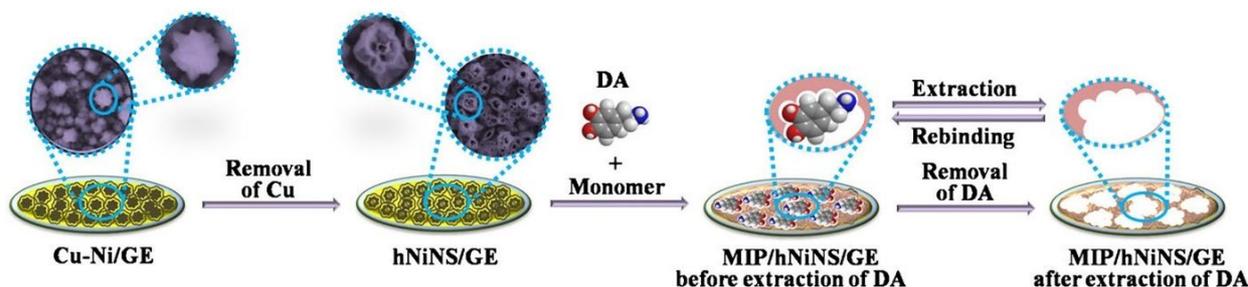


Fig. 1. Schematic representation of preparing MIP/hNiNS/GE. Reproduced from Liu *et al.*, 2017, with permission from *Microchimica Acta* [28].

40 μM in PBS (pH 7.0). The sensor could successfully determine the concentrations of DA in human urine samples with the recoveries of *ca.* 97.6-102.6% [31].

A conducting polymer-modified electrode was proposed in the research by Hesieh and Whang, which was fabricated by electropolymerization of 5-amino-1H-tetrazole (ATet) on a GCE. Electrochemical studies such as DPV and CHA were performed for the evaluation of the rate constant of the catalytic reaction ($9.69 \times 10^3 \text{ M}^{-1} \text{ s}^{-1}$), the diffusion coefficient of the analyte DA ($7.35 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$) and the linear dynamic range of the DA determination (5.0 μM -1.0 mM). The film modified electrode has superior resolving power in quantitative determination from the mixture of analyte and it was found to be an efficient functionalized electrode for its sensitivity and selectivity toward the analyte of interest [32].

The research by Manikandan *et al.* is based on the use of a redox mediator containing an azo group for the selective determination of DA in the presence of UA and AA. A modified electrode was prepared by electrochemical polymerization of the poly 2-naphthol orange film (P2NO) on the paraffin wax-impregnated graphite electrode (PIGE) by applying potential between -0.6 and 0.8 V at scan rate of 50 mV s^{-1} for 30 segments. The P2NO film modified electrode exhibited selective determination of DA in the presence of UA and AA and the electrocatalytic activity for oxidation of DA was excellent. The linear range for the determination of DA was 0.6 to 250 μM with a limit of detection of 0.13 μM [33].

A polymerized film of copper-2-amino-5-mercapto-1,3,4-thiadiazole (Cu(II)-AMT) complex (poly (Cu-AMT)) was synthesized by Li *et al.* (Fig. 2). The biomimetic sensor

enables a reliable and sensitive determination of DA with a linear range of 0.01-40 μM and a detection limit of 3.48 nM at a signal-to-noise ratio of 3 [34].

A sensitive electrochemical sensor was made by the electro polymerization of L-cystine (L-Cys) on the surface of a GCE followed by drop casting of MWCNTs reported by Shaikshavali *et al.* The sensor showed good electrocatalytic activity towards the sensing of DA and simultaneously discriminated DA in presence of UA and FA. The effect of concentration on the DA was studied and a linear relationship was observed between the concentration 10.0-200.0 μM , with a limit of detection and limit of quantification values as $2.8 \times 10^{-6} \text{ M}$ and $9.6 \times 10^{-6} \text{ M}$ [35].

Dopamine Determination by Modified Electrode with Graphene and Graphene Oxide

The 2D hexagonal lattice graphene emerged as an ideal candidate who has captured enormous interests and grown explosively in electrocatalysis and electrochemical biosensors. The sp^2 hybridization, unique electronic configuration, its extraordinary electron transport property, so called ballistic conductivity, and the number and position of the oxygen functionalities of graphene helps to promote and accelerate the electron transfer process during electrochemical detection. However, the ability of rapid electron transfer kinetics at the basal planes of graphene sheets act as nanoconnectors and thus electrically facilitates the heterogeneous electron transfer between the biomolecules and electrode substrate, resulting in excellent sensing performance.

A screen-printed electrode was modified by graphene quantum dots (GQD/SPE) in order to be used as sensor for

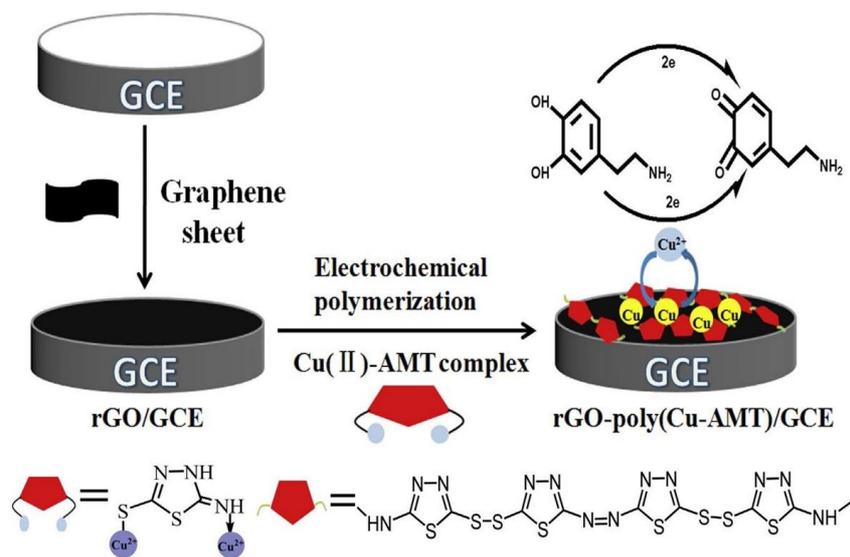


Fig. 2. The fabrication process of the biomimetic sensor and the catalytic mechanism for the analysis of dopamine. Reproduced from Li *et al.*, 2017, with permission from Talanta [34].

DA in the presence of tyrosine by Beitollahi *et al.* To evaluate the efficiency of the developed electrode toward detection of dopamine and tyrosine in aqueous solutions, various electrochemical methods including CV, CHA and DPV techniques were employed. Application of GQD/SPE created a separation of 435 mV in the oxidation peak potentials of DA and tyrosine. The calibration curves was within the range of 0.1-1000.0 and the detection limit (S/N = 3) was 0.05 μM for DA. The diffusion coefficients using chronoamperometry at the surface of modified electrode was determined as $9.0 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$ [36].

Mahbubur *et al.* developed a graphene nanoplatelet modified fluorine doped tin oxide electrode (GNP/FTO) for the simultaneous detection of DA and UA in the presence of AA and investigated the interaction mechanisms of DA, UA, and AA with GNPs considering their charging states at different pH values. Owing to the unique structure and properties originating from the oxygen and nitrogen functional groups at the edges, GNPs showed high electrocatalytic activity for the electrochemical oxidations of AA, DA, and UA with peak to peak potential separations (ΔEP) between AA-DA and DA-UA of *ca.* 0.23 and 0.17 V, respectively. These values are sufficiently high to allow the simultaneous detection of DA and UA without interference

from AA. The highly sensitive and stable GNP/FTO sensor showed sensitivity of *ca.* $0.15 \pm 0.004 \mu\text{A}/\mu\text{M}$ with detection limit of *ca.* $0.22 \pm 0.009 \mu\text{M}$ for DA [37].

Yao *et al.* described an amperometric sensor for DA which has a working potential as low as +0.02 V (*vs.* SCE). It makes use of a reduced graphene oxide (rGO) and manganic manganous oxide (Mn_2O_4) in a film of Nafion on gold nanoparticles deposited on a gold electrode. The electrochemical properties of the modified electrode were investigated by CV and electrochemical impedance spectroscopy (EIS) and amperometric methods. After method optimization, the amperogram displays a linear range extending from 1.0-1.45 mM with a limit of detection as low as 0.25 μM (at an S/N ratio of 3). The modified electrode was employed for the determination of DA in injection solution samples with satisfactory results [38].

Tian *et al.* described a voltammetric sensor for simultaneous determination of DA, UA, L-tyrosine (Tyr) and the diuretic drug hydrochlorothiazide (HCTZ). The assay is based on the use of graphene nanowalls deposited on a tantalum substrate. The graphene nanowalls display a large specific surface, high conductivity, and a large number of catalytically active sites for oxidation of analytes. Simultaneous detection is performed best at a pH value of

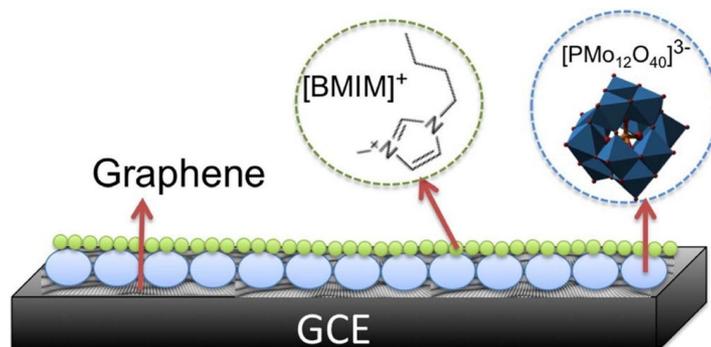


Fig. 3. The structure of ionic liquid-phosphomolybdic acid/graphene modified electrode. Reproduced from Zhao *et al.*, 2017, with permission from ECS Journal of Solid State Science and Technology [44].

7.0 and at peak potentials of 0.124 V (*vs.* SCE) for DA. The respective detection limit was 0.04 μM and linear calibration for DA was obtained over the range of 0.3-40.0 μM [39].

Kim *et al.* reported the determination of DA and AC using activated graphene-Nafion modified glassy carbon electrode (AG-NA/GCE). The electrochemical characterization of AG-NA/GCE was examined with CV analysis of potassium ferricyanide. The electroanalytical performance of AG-NA/GCE toward the oxidation of DA and AC was analyzed via CV and DPV. The results showed excellent selectivity and sensitivity analyses for DA with low detection limit value of 0.33 μM ($S/N = 3$). As for the examination and evaluation of electrochemical activities in real samples, the determination of DA and AC was examined with AG-NA/GCE in diluted human urine samples [40].

Jahani and Beitollahi reported an electrochemical method based on a disposable graphene nanosheets and NiO nanoparticles modified carbon screen printed electrode (NiO/GR/SPE) to describe the simultaneous determination of DA and UA. A peak potential difference of 150 mV between DA and UA was large enough to determine DA and UA individually and simultaneously. The anodic peak currents of DA were found to be linear in the concentration range of 1.0-500.0 μM with the detection limit of 3.14×10^{-7} M [41].

Lue *et al.* describe a three-dimensional (3D) structure composed of graphene and DNA for use in a voltammetric dopamine (DA) sensor. The modified gold electrode possesses excellent electrocatalytic activity for

determination of DA, best at a working voltage of 0.1 V (*vs.* SCE). Response to DA is linear in the 0.1 to 100 μM concentration range, with a 30 nM detection limit [42].

Thaleb *et al.* describe the fabrication of an interconnected edge-exposed graphene nanostructure *via* chemical vapor deposition of foliated graphene onto a network of alumina nanofibers. The fibers obtained were shown to enable voltammetric determination of DA, UA and AA. Under optimum conditions, the calibration plot is linear in the 1-80 μM range for DA with detection limit of 0.47 μM . The sensor was successfully applied to the simultaneous determination of DA and UA in the presence of AA in spiked urine sample [43].

A reduced graphene oxide (RGO) coated glassy carbon electrode modified with ionic liquid (IL, [BMIM] [BF₄]) and phosphomolybdic acid (PMo₁₂, H₃PMo₁₂O₄₈) was fabricated by (IL-PMo₁₂/RGO/GCE) Zhao *et al.* The phosphomolybdic acid anionic monolayer could electrostatically adsorb ionic liquid cations to form an organic and inorganic hybrid film on graphene sheets (Fig. 3). The modified electrode was used for the determination of DA in the presence of UA. The linear range of the modified electrode was from 0.1-100 μM for determination of DA with an R-Square 0.9924 and with a detection limit of 3.3×10^{-8} M ($S/N = 3$). The IL-PMo₁₂/RGO/GCE also presented good stability and reproducibility [44].

Park *et al.* fabricate an electrochemical sensor by modifying an electrode with reduced graphene oxide sheet (rGS)-gold nanoparticle (GNP) complexes to determine DA and AA in a mixture within a range of 0.1-100 μM . The rGS

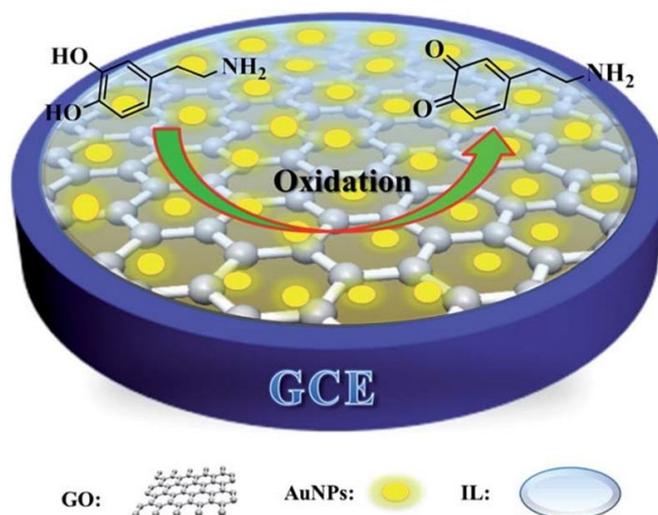


Fig. 4. Schematic illustration of the electrochemical sensors for the determination of DA. Reproduced from Li *et al.*, 2017, with permission from RSC Advances [46].

was used to improve the selectivity of the sensor by having π - π interaction with DA, and the GNPs improved the sensitivity due to their great conductivity and large surface area. The results showed that it had linearity in the range of 0.1-100 μM of DA and the limit of detection was 0.098 μM in the presence of AA 400 μM . Therefore, it could be applied to medical biosensors to diagnose neural diseases [45].

An electrochemical sensor for detection of DA has been developed based on ionic liquid functionalized graphene oxide supported gold nanoparticles (GO-IL-AuNPs) coated onto a GCE, suggested by Li *et al.* (Fig. 4). The activities of hybrid nanomaterials toward the oxidation of DA were analyzed *via* CV and DPV. Under the optimum conditions, DPV was employed to detect ultra-trace amounts of DA, for which a wide linear range of 7 nM-5 mM and a limit of detection 2.3 nM ($S/N = 3$) were obtained. The proposed sensor exhibited excellent accuracy and precision and the relative standard deviation (RSD) was less than 5% [46].

Yue and Kim described the fabrication of an electrode modified with electrochemically RGO for the simultaneous electrocatalytic detection of DA. RGO was formed on a GCE by the reduction of graphene oxide (GO) using linear sweep voltammetry. The RGO/GCE was formed by

subjecting a GO solution (1 mg ml^{-1} in 0.25 M NaCl) to a linear scan from 0 V to -1.4 V at a scan rate of 20 mV s^{-1} . The detection limit for DA was found to be 0.46 μM , using the amperometric *i-t* curve technique, with the S/N ratio being 3 [47].

Dopamine Determination by Modified Electrode with Carbon Nanotubes

Carbonaceous materials have received considerable interest to be used as electrode materials owing to their low cost, good electron transfer kinetics, and biocompatibility. Carbon nanotubes (CNTs) are viewed as rolled version of graphene sheets which are one of the exciting materials for fabricating electrochemical biosensors. CNTs have shown good electrocatalytic properties in the oxidation and reduction of a wide variety of compounds that can be attributed to their unique properties including enhanced electronic properties, large edge lane/basal plane ratio, outstanding mechanical strength, and rapid electrode kinetics. An electrochemical sensor for the simultaneous determination of DA, AC, folic acid (FA) and N-acetylcysteine (NAC) is described by Beitollahi *et al.* The sensor is based on CPE modified with 5-amino-3',4'-dimethyl-biphenyl-2-ol (5ADB) and takes the advantages of carbon nanotubes (CNTs), making the modified electrode

highly sensitive for the electrochemical detection of these compounds. Under the optimum pH of 7.0, the oxidation of DA occurs at a potential about 170 mV less positive than that of the unmodified CPE. The results of this study indicated that the electrode exhibited linear response over a wide concentration range 1.2-900.0 μM with a detection limit of 0.57 μM [48].

A DA electrochemical sensor was fabricated based on magnetism-assisted modification of SPE with magnetic multi-walled carbon nanotubes (mMWCNTs) suggested by Zhang *et al.* The linearity was found between 5-180 μM , with the detection limit of 0.43 μM . This electrode has been applied to determine DA concentration in spiked human blood serum and satisfactory recoveries were found in the range of 97.43-102.94% with the RSDs of less than 2.27% [49].

A sensor was developed by Nagles *et al.* in order to determine DA and UA by adsorptive voltammetry, using optimal amounts of single walled carbon nanotubes (SWCNT) dispersed in chitosan solution (cs) and deposited on a SPCE. The electrode surface (cs-SWCNT/SPCE) was treated with the ionic liquid (IL) 1 butyl-3-methylimidazolium tetrafluoroborate (BMIMBP₄). The electrocatalytic properties of cs-SWCNT-IL/SPCE were studied with CV and electrochemical EIS. Anodic peak currents for DA and UA with BMIMBP₄ in the presence of ascorbic acid (AA) increased by 17.0% and 70.0%, respectively. Anodic peak currents were proportional to the concentration between 0.5 and 5.0 μM using standard solutions of DA and 0.50-30.0 μM for DA with real samples. Detection limit was 0.16 μM . The sensor was used in the determination of DA in human urine samples spiked with quantities of DA with recoveries between 85 and 102% for DA [50].

In another work, unzipped multiwalled carbon nanotube oxides (UMCNOs) was obtained from longitudinally unzipped multiwalled carbon nanotubes (MWCNTs) by the Hummers method. Then, overoxidized poly(*p*-aminophenol) (pAPox) and UMCNOs copolymer modified electrode was proposed for the simultaneous determination of DA, UA and tryptophan (TRP). Besides, the coexistence of AA has no interference toward the detection of DA, UA and Trp. Under the optimal conditions, linear calibration for DA was obtained over range of 15.0-515.0 μM with detection limit

(S/N = 3) of 0.33 μM [51].

Beitollahi *et al.* investigated for the simultaneous determination of DA, UA and TRP using a multiwall carbon nanotubes/5-amino-3',4'-dimethoxy-biphenyl-2-ol modified carbon paste electrode (5ADMBCNPE). The 5ADMBCNPE displayed excellent electrochemical catalytic activities towards the oxidation of DA, UA and TRP. The electrochemical profile of the proposed modified electrode was analyzed by CV that showed a shift in the oxidation peak potential of DA at 160 mV to a less positive value compared with an unmodified carbon paste electrode. SWV in 0.1 M PBS at pH 7.0 was performed to determine DA in the range from 1.2-800.0 μM , with a detection limit of 0.16 μM . The present method was applied to determine DA in some real samples [52].

The redox response of a modified carbon nanotube paste electrode of 2,2'-[1,2-ethanediylbis (nitriolethylidene)]-bis-hydroquinone was investigated by Mazloum Ardakani *et al.* Mixture of DA and UA can be separated from one another with a potential difference of 180 mV between them at a scan rate of 25 mV s^{-1} by cyclic voltammetry. These conditions are sufficient to allow determination of DA and UA both individually and simultaneously. The electrocatalytic current increases linearly with the DA concentrations in the range of 0.1-900 μM and the detection limit of 0.087 μM . The diffusion coefficient (D ($\text{cm}^2 \text{s}^{-1}$) = 7.3×10^{-6}) and the kinetic parameters such as the electron transfer coefficient, ($\alpha = 0.32$) and the heterogeneous rate constant, (k' (cm s^{-1}) = 2.21×10^{-3}) for DA were determined using electrochemical approaches [53].

MWCNTs/MoS₂ decorated Cobalt oxide polyhedrons (MWCNTs/MoS₂/Co₃O₄) were synthesized *via* facile hydrothermal route reported by Sakthivel *et al.* An electrochemical non enzymatic DA assay was described using MWCNTs/MoS₂/Co₃O₄ PHs modified SPCE. The sensor displayed outstanding sensitivity with nanomolar detection limit of 0.013 μM and a linear calibration for DA was obtained over the range of 2150.2-5540 μM [54].

Beitollahi and Sheikhshoae fabricated a carbon paste electrode modified with a molybdenum(VI) complex and carbon nanotubes and apply it for the electrocatalytic oxidation of DA that reduced the overpotential by about 125 mV with obviously increase in the current

response. Due to its strong electrocatalytic activity towards DA, the modified carbon paste electrode can resolve the overlapped voltammetric waves of DA and AC into two well-defined voltammetric peaks with peak to peak separation in a potential of about 230 mV. In phosphate buffer solution of pH 7.0, the oxidation current increased linearly with two concentration intervals of DA, one is 0.1-40.0 μM and, the other is 40.0-800.0 μM . The detection limit (3σ) obtained by DPV was 76.0 nM. The proposed method was successfully applied to the determination of DA, and AC in some commercial pharmaceutical samples [55].

Modification of gold electrode using SWCNTs has been done by Kurniawan *et al.* The synthesis of SWCNTs on the gold electrode surfaces was carried out using chemical vapor deposition method. The performance of the electrodes as a sensor was characterized using DA solution at pH 4 by CV. The detection limit and sensitivity of the electrodes are 0.79 μM and 3.414 $\mu\text{A mm}^{-2} \mu\text{M}^{-1}$. No interference signal was found from AA during DA measurement [56].

Graiti *et al.* demonstrate a probe-style electrochemical sensor using highly flexible and conductive multi-walled carbon nanotubes (MWNT) yarns. The MWNT yarn-based sensors can be fabricated onto micro Pt-wire with a controlled diameter varying from 100 to 300 μm , and then further modified with Nafion *via* a dip-coating approach. The fabricated micro-sized sensors were characterized by electron microscopy, Raman, FTIR, electrical and electrochemical measurements. For the first time, the MWNT/Nafion yarn-based probe sensors were assembled and assessed for high-performance DA sensing that showed a significant improvement in both sensitivity and selectivity in DA detection in presence of AA and UA. It offered a potential for further development as implantable probe sensors [57].

Vidiya and Kumara Swamy used carbon nanotubes/Tween-20 for the modification of CPE. In the presence of potassium ferrocyanide, the modified CPE showed a redox peak in 1M KCl solution as a supporting electrolyte. The modified electrode also showed good electrocatalytic activity towards DA in 0.1M phosphate buffer solution at pH 7.4, and the detection limit of DA was found to be 0.27 μM [58].

Dopamine Determination by Modified Electrode with Nanocomposites

Electrocatalysis at nanocomposite based modified electrodes offer significant advantages for the determination of many drugs and bio-substances. These electrodes can enhance the electron transfer rate and reduce the overpotential of the oxidation of many substances. Therefore, these modified electrodes can be used as electrochemical sensors in clinical, medical, industrial, environmental and agricultural analyses. Beitollahi *et al.* reported construction of GO/Fe₃O₄@SiO₂ core-shell nanocomposite modified graphite screen printed electrode that showed advantageous characteristics in terms of electroconductivity, catalytic activity and surface area compared to bare screen printed electrode. The designed sensor was used for determination of DA and UA. Using modified electrode DA was determined in the concentration range of 0.1-600.0 μM , whereas the detection limit was found to be 8.9×10^{-8} M. Real sample analysis was done successfully using modified electrode [59].

A carbon paste electrode modified with ZnO nanorods and 3-(4'-amino-3'-hydroxybiphenyl-4-yl)-acrylic acid (3, 4'-AAZCPE) was fabricated by Mollakbari *et al.* It was found that oxidation of DA at the surface of modified electrode occurs at a potential of about 180 mV less positive than that of an unmodified carbon paste electrode. The linear range of modified electrode was from 0.3-100.0 μM and the detection limit of 5.6×10^{-8} M for DA was obtained using SWV. This modified electrode was used for simultaneous determination of DA, melatonin, methionine and caffeine. The modified electrode was used for determination of DA, melatonin, methionine and caffeine in human blood serum and urine samples [60].

An electrochemical sensor was prepared by the modification of CPE with a synthesized nanocomposite reported by Sheikh-Mohseni and Pirsá. The structure and morphology of the nanocomposite prepared by combination of poly pyrrole and copper oxide nanoparticles (PPy/CuO) was studied. This nanocomposite showed a good electrocatalytic activity; so that the nanocomposite based electrochemical sensor (CPE-PPy/CuO) exhibited an excellent electroanalysis signals towards oxidation of DA and AC. The calibration plot for DA had linear segment

Table 1. Some Analytical Performances Attained in DA Electrochemical Determination

Electrode	Electrochemical method of determination	Modifier	Modification method	Linear range	Detection limit	Simultaneously used analytes	Ref.
CPE	SWV	QDs	Bulk	7.5×10^{-8} - 6.0×10^{-4} M	$(2.1 \pm 0.1) \times 10^{-8}$ M	UA	[68]
CPE	CV, DPV	CoL	Bulk	1×10^{-5} - 1×10^{-3} M	2×10^{-6} M	AA	[69]
CPE	CV, DPV	CoClSal	Bulk	2×10^{-6} - 1×10^{-3} M	5×10^{-7} M	AA	[70]
CPE	CV	EDC.HCl/ HOBt	Bulk	0.569-515 μ M	0.190 μ M	H ₂ O ₂	[71]
CPE	CV	BzTC	Bulk	10-60 μ M	-	AA	[72]
CPE	DPV	GR/ MoS ₂	Bulk	5.0×10^{-8} - 1.0×10^{-5} M	7.13×10^{-9} M	-	[73]
CPE	CV, DPV	WO ₃ NPs	Bulk	0.1-1.7 μ M	0.58 μ M	UA, FA	[74]
GCE	CV, DPV	3DGH- AuNPs	Surface	0.2-30 μ M	2.6 nM	AA, UA	[75]
GCE	CV, DPV	RGO/TiN	Surface	0.1-80 μ M	0.012 μ M	AA, UA	[76]
SPE	CV, DPV	GO/Fe ₃ O ₄ @SiO ₂	Surface	0.1-600.0 μ M	8.9×10^{-8} M	UA	[77]
GCE	CV, DPV	RGO/AuN Ps	Surface	0.1-100.0 μ M	0.14 μ M	AA, UA	[78]
CPE	CV	Co ₃ O ₄ /Cu O/SDS	Surface	1.0-80.0 μ M	0.6 μ M	AA, UA	[79]
ITO	CV	RGO/AuN Ps	Surface	0.1-30 μ M	1.28 μ M	AA, UA	[80]
GCE	CV, DPV	SnO ₂ /chito san	Surface	1-18 μ M	0.77 μ M	AA, UA	[81]

Table 1. Continued

SPE	CV, DPV	N/QDs/chitosan	Surface	1.0-200 μM	0.145 μM	AA, UA	[82]
ITO	CV, DPV	RGO/AuNPs	Surface	1.0-100 μM	0.6 μM	-	[83]
SPE	CV, DPV	GR/GLN	Surface	0.05-79.5 μM	0.0045 μM	-	[84]
GCE	CV, DPV	RGO/poly (L-lysine)	Surface	2.0-60 μM	0.10 μM	AA, UA	[85]
GCE	CV, DPV	Poly(β -cyclodextrin)/QDs	Surface	4-220 μM	0.14 μM	UA, TRP	[86]
GCE	DPV	Poly (Dianix Yellow)	Surface	0.035-2.5 μM	4.64 nM	AA, UA	[87]
GCE	CV, SWV	NiHCF/PDAN	Surface	600-1000 μM	0.034 μM	AA, UA	[88]
GPE	LSV	PTh	Bulk	10-180 μM	1 μM	UA, TRP	[89]
GCE	CV	Poly ferulic acid/MWCNT	Surface	5-120 μM	2.2 μM	NADH, EP	[90]
GCE	CV	MWCNTs/WO ₃	Surface	0.02-0.5 mM	0.018 μM	AA	[91]
CPE	CV, DPV	poly ferric sulfate/CNT	Bulk	0.8-261 μM	0.2 μM	-	[92]
GCE	CV	Hexacyanoferrate /GR	Surface	0.4-28 μM	0.03 μM	UA	[93]
GCE	CV, DPV, LSV	GO-BAMB-Co(OH) ₂	Surface	3-100 μM	0.4 μM	UA	[94]
CF	CV	RGO/QDs	Surface	0.1-100 μM	0.02 μM	AA, UA	[95]
GCE	CV, DPV	GA-RGO/AuNPs	Surface	0.01-100.3 μM	2.6 nM	AA, UA	[96]

including 0.060-1000.0 μM and detection limit was found to be 0.020 μM . The electrochemical sensor was used for determination in real samples [61].

One kind of nanocomposite consisting of graphene and polypyrrole was synthesized *via* a facile and mild way with the assistance of microwave irradiation suggested by Rui *et al.* Dopamine sensor based on the composite showed a sensitivity of $363 \mu\text{A mM}^{-1} \text{cm}^{-2}$, a linear range of 1×10^{-4} - 1×10^{-3} M, and a detection limit of 2.3×10^{-6} M (S/N = 3) [62].

A nanocomposite comprising RGO and titanium nitride (TiN) was fabricated by Haldorai *et al.* Transmission electron microscopy showed that TiN nanoparticles with a mean diameter of < 10 nm were densely decorated onto the RGO surface. CV indicated that a GCE modified with the RGO-TiN nanocomposite has an excellent electrocatalytic activity towards the oxidation of DA in 0.1 M phosphate buffer solution (pH = 7). The combined use of RGO and TiN resulted in a higher current response and a lower oxidation potential (0.23 V) than those of bare GCE, RGO/GCE and TiN/GCE. The developed electrochemical sensor showed a linear relationship with the DA concentration from 0.1 to 80 μM with a detection limit of 0.012 μM (S/N = 3). The sensor was applied for the determination of DA in urine samples by the standard addition method and gave recoveries of 97.0-101.5% [63].

A hybrid graphene-ZIF-8 (G-ZIF-8) nanocomposite modified electrode was prepared in research by Zheng *et al.* G-ZIF-8 modified electrode exhibited excellent electroanalytical performance for DA. The linear concentration range was from 3.0-1.0 mM with the detection sensitivity of 0.34 mA/mM and the detection limit of 1.0 mM was obtained. The prepared sensor was applied to the detection of DA in serum sample with recoveries from 96.8% to 100.7% [64].

A novel SnO_2 /chitosan (CHIT) nanocomposite modified GCE has been successfully fabricated by Selvarajan *et al.* for simultaneous determination of DA, AA and UA *via* CV and DPV. DPV studies showed that there is a linear relationship between the peak current and concentration in the concentration range of 1-18 μM for DA with the detection limit (S/N = 3) of 0.77 μM . The proposed method was applied for the simultaneous determination of AA, DA and UA, in real samples [65].

A new modification strategy was reported by Ganjali *et al.* to modify a GCE based on $\text{ZnO}/\text{Al}_2\text{O}_3$ nanocomposite. This modified electrode was designed in order to be used as a sensitive and selective sensor towards detection of trace amount of DA. The effective parameters on the optimal performances of the electrode such as pH of the test solution and the applied scan rate during the electrochemical process were also studied. This sensor responded linearly towards detection of DA within a wide range of 5.0×10^{-6} - 7.0×10^{-4} M with a low detection limit of 2.0×10^{-6} M (pH = 7.0), under the optimum conditions [66].

An electrochemical sensor for determining DA was developed by modifying phytic acid/graphene oxide (PA/GO) nanocomposites onto a GCE. PA functionalized GO was prepared by an ultra-sonication method. The proposed electrochemical sensor was applied to detect various concentrations of DA by DPV. The PA/GO/GCE was considered to be highly sensitive to DA in the range of 0.05-10 μM . The PA/GO/GCE demonstrated high electrochemical selectivity towards DA in the presence of AA and UA [67].

CONCLUSIONS

Tremendous efforts have been made over the last thirty years to detect biogenic amines, especially dopamine. Information regarding the temporal fluctuations of the dopamine concentration in the brain is critical for understanding its widespread effects as a neurotransmitter. Moreover, abnormal dopamine transmission has been linked to several neurological disorders, *e.g.*, schizophrenia, Huntington's disease and Parkinson's disease. As parkinson's disease is characterized by a severe depletion of the dopamine pool, the ability to sensitively and selectively measure the concentration of the neurotransmitter dopamine could potentially be used for molecular diagnosis of Parkinson's disease. In this review, we have summarized the efforts made in electrochemical detection of dopamine with high sensitivity and selectivity by modifying the electrode surface with different modifiers.

Analytical Performances of Electrochemical DA Sensors

The analytical performances of electrochemical methods

depend on the sensor's construction. Some of the most illustrative examples are extensively reviewed in Table 1.

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