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REVIEW ARTICLE

Carbon Paste Modified Electrode as Powerful Sensor Approach Determination of Food Contaminants, Drug Ingredients, and Environmental Pollutants: A Review

Hassan Karimi-Maleh[a,b,*], Fatemeh Karimi[a], Morteza Rezapour[c], Majede Bijad[d], Mohammad Farsi[d], Aliasghar Beheshti[e] and Seyed-Ahmad Shahidi[f]

[a]*Department of Chemical Engineering, Laboratory of Nanotechnology, Quchan University of Technology, Quchan, Iran;* [b]*Department of Applied Chemistry, University of Johannesburg, P. O. Box 17011, Doornfontein Campus, 2028, Johannesburg, South Africa;* [c]*IP Department, Research Institute of Petroleum Industry (RIPI), P.O. Box 14665-137, Tehran, Iran;* [d]*Department of Food Science, Sari Branch, Islamic Azad University, Sari, Iran;* [e]*Department of Water Resources Engineering, Ferdowsi University of Mashhad, Mashhad, Iran;* [f]*Department of Food Science and Technology, Ayatollah Amoli Branch, Islamic Azad University, Amol, Iran*

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Abstract: ***Background*:** Application of electrochemical sensors for analysis of food, biological and water polluting compounds helps to speed up their analysis in the real samples. Electrochemical sensors with low cost, fast response and portable ability are a better choice compared to traditional methods for analysis of electro-active compounds such as HPLC. Therefore, in recent years, many analytical scientists have suggested this type of analytical method for analysis of food, biological compounds and water pollutants.

***Objective*:** Due to low cost, easy modification and low non-faradic current, the carbon paste electrode is a suitable choice as a working electrode in the electrochemical and especially voltammetric analysis. On the other hand, modification of carbon paste electrode can improve the quality of the sensor for the analysis of electroactive compounds at nanomolar level.

Keywords: Carbon paste electrode, drug and water pollutant analysis, electrochemistry, food, modified electrode, electrochemical sensors.

1. INTRODUCTION

The analysis of food, drug, biological and water polluting compounds is very important for human health [1-10]. Due to the significant growth of harmful additives such as azo dyes and synthetic antioxidants in food products, the need for the detection and measurement of food additives has increased [11-16]. Also, to determine the level of permitted additive such as ascorbic acid and folic acid in food products is very important due to their control in the human body [17-19]. In addition, analysis of drug and biological compounds such as glutathione, cysteine, homocysteine, anticancer drugs, morphine *etc*. in biological samples such as urine and blood is very important for checking the human health [20, 21]. In between, change in the level of some biological compounds such as glutathione, homocysteine and uric acid in the body can be a serious danger for the human body.

As an example, the increase in the homocysteine level as an amino acid can be relative to heart disease [22]. The change in the level of homocysteine in the human body can be relative to vitamins B_{12}, folate and B_{12} concentration [23].

As an example, the increase in the homocysteine level as an amino acid can be relative to heart disease [22]. The change in the level of homocysteine in the human body can be relative to vitamins B_{12}, folate and B_{12} concentration [23], which can be a sign of problems in the immune system. On the other hand, the reduction in glutathione concentration can increase the risk of disease for humans [24]. In addition, the overdose of some drugs can be harmful to the human body. As an example, the increase in acetaminophen level can be toxic for the body. Moreover, vomiting, diarrhea, loss of appetite, abdominal pain or cramping irritability and sweating are the main effects of overdose of acetaminophen [25].

In addition to this, attention to water pollutants is an important point for the investigation of drinking water quality [26-28]. The transmission of many diseases through water has caused the analysis of water pollutants to become an important and challenging topic [29].

According to the above points, the necessity for the biological analysis of, drug, food and water pollutants compound in real samples has increased. Some analytical

*Address correspondence to this author at the Department of Chemical Engineering, Laboratory of Nanotechnology, Quchan University of Technology, Quchan, Iran; Tel: +98-911-254-0112;
E-mail: h.karimi.maleh@gmail.com

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methods such as HPLC, gas chromatography, spectroscopy, flow injection analysis, NMR, chemiluminescence, liquid-liquid microextraction, ICP and electrochemical sensors were suggested for the biological analysis of, drug, food and water pollutants in the real samples such as water, blood, urine, fruit juices, ampule and tablet [30-50].

During the analytical method, electrochemical sensors can be selected as fast, sensitive and portable systems with good selectivity for analysis of biological, gas pollutants, drug, food and water pollutants compounds [51-60]. On the other hand, the variety of electrochemical methods can be useful for the analysis of different types of compounds.

Voltammetric methods are sensitive, selective and fast electrochemical techniques and a good choice for the analysis of electro-active compounds [61-65]. A voltammetric method needs three electrodes including references electrode, counter electrode and working electrode. The working electrode plays the main role in an electrochemical cell. There are different types of working electrodes such as Au electrode, Pt electrode, mercury electrode, glassy carbon electrode, pencil graphite electrode, Ni foam, quartz crystal microbalance electrode, boron doped diamond paste electrode, carbon paste electrode *etc*. [66-70]. The application of Au electrode, Pt electrode, mercury electrode, glassy carbon electrode, pencil graphite electrode, Ni foam, quartz crystal microbalance electrode, and boron doped diamond paste electrode caused many problems compare to carbon paste electrode. Their modification and renewability are very hard as compare to carbon paste electrodes [71].

Prof. Ralph N. Adams introduced the carbon paste electrode in 1958 as a new working electrode in a paper entitled "Carbon Paste Electrodes" [72]. He suggested a coupling of pure graphite powder and liquid binder for the fabrication of carbon paste electrodes. The carbon paste electrode and mercury electrode were introduced at the same time as a working electrode in electrochemical systems.

Due to high anodic windows compare to mercury electrode, the application of this type of electrode has increased as new electrochemical sensors in the anodic voltammetric analysis [73]. In between 1958-1963, the carbon paste electrode was used for the investigation of electroactive redox mechanism and other qualitative studies. In addition, a few papers suggested carbon paste electrode as a sensor for analysis of electroactive compounds. For example, Olson *et al*. used carbon paste electrode for the study of oxidation and reduction of p-nitroaniline [74].

The N-methylaniline and N,N-dimethyl-p-Toluidine oxidation mechanisms were investigated at a surface of platinum and carbon paste electrodes with voltammetric methods by Galus and Adams [75]. Jacobs introduced the carbon paste electrode as an analytical sensor for the determination of gold and silver ions [76]. After 1964, due to low electrical conductivity and low selectivity of carbon paste electrode, the efforts to modify the carbon paste electrode were undertaken. Many scientific reports described the applications of different modifiers for improving sensitivity and selectivity of carbon paste electrodes [77-80]. In between, nanomaterials such as carbon nanotubes, metal oxide nanoparticles, graphene and nanocomposites showed good improvement in the electrical conductivity of carbon paste electrodes [81-84]. In addition, the biomolecules such as DNA and enzyme showed powerful ability for modification of carbon paste electrodes as for fabrication electrochemical biosensors [85-87]. In recent years, the replacement of nonconductive binders such as paraffin oil or nujol with room temperature ionic liquids as conductive binders improved the ability of carbon paste electrode for nanomolar analysis of electroactive compounds. Also, simultaneous modification of carbon paste electrode with room temperature ionic liquids and nanomaterials showed good sensitivity for the biological fabrication of analytical sensors in food, drug and water pollutants compounds analysis [88-93].

In conclusion, carbon paste modified electrodes can be suggested as powerful tools for the voltammetric analysis of food, biological and water polluting compounds. The modification of carbon paste electrodes improved the selectivity and sensitivity of the sensors for trace level analysis.

2. ADVANTAGE OF MODIFICATION OF CARBON PASTE MODIFIED ELECTRODES

Modification of carbon paste electrode is the main subject of the present study. Therefore, we would like to describe important modification of carbon paste electrodes. Two factors are effective on the carbon paste electrode signals in the presence of electroactive compounds.

1) Powder purity (as conductive matter and base of the electrode)
2) Liquid binders

There are two methodsfor the modification of a carbon paste electrode. In the firstmethod, a conductive powder such as multi-wall carbon nanotubes, single wall carbon nanotubes, graphene, metal oxide nano-powders *etc*. was added. to carbon paste matrix [94-97]. This comes to mind, why all of the graphite powder cannot be replaced with conductive materials such as multiwall carbon nanotubes?. Large amount of nanomaterials increased the non-faradaic current and removed the oxidation/reduction signals of electro-active compounds. Therefore, the ratio of graphite powder and conductive materials must be optimized in the carbon paste matrix as an important factor for the modification of carbon paste electrode. In addition, carbon paste electrode is an exceptional choice for the modification of the working electrode with electro-active mediators. As we know electro-active mediators such as ferrocene and its derivatives, inorganic complexes, catechol derivatives *etc*. can be useful for increasing the selectivity of electrochemical sensors due to their electro-catalytic interaction ($EC^{/}$ mechanism) with electro-active materials. The incorporation of this type of mediators into carbon paste electrode is easier as compared to solid electrodes [98-101]. In the second way, the carbon paste electrode can be modified with a conductive

binder. Usual binders such as paraffin oil and nujol are nonconductive because they have aliphatic hydrocarbons structures. But, many years ago, they were used as a binder for fabrication of carbon paste electrodes. In the recent years, room temperature ionic liquids were suggested as a conductive binder for the fabrication of carbon paste electrodes [102-110]. Due to the ionic structure of ionic liquids, they are a good choice as a binder for fabrication of carbon paste electrodes. Due to the high viscosity of room temperature ionic liquids, the high value of this binder can increase the capacitive current in voltammetric investigations. The room temperature ionic liquids athigh level can cover the electrode surface and exchange of electrons of a redox system with electrode surface (electroactive sites of carbon) will be hard. Therefore, the ration of nonconductive binder to room temperature ionic liquid must be optimized before the starting of electro-active compound analysis. The carbon paste electrode has some advantage compared to Au electrode, Pt electrode, mercury electrode, glassy carbon electrode, pencil graphite electrode, Ni foam, quartz crystal microbalance electrode, boron doped diamond paste electrode,) such as low background currents, simple renewability, simple modification and specific reaction kinetics [111-115].

Here, we are going to consider the reported papers and their advantages for using carbon paste modified electrodes as a sensor for food, biological and water pollutants compound analysis.

3. MODIFIED CARBON PASTE ELECTRODE AS A FOOD SENSOR

The analysis of food compounds and especially food additives is important in food products due to the direct relation between food safety and human health. The presence of forbidden additives in food products can increase the risk of cancer. Addition of forbidden additive in food samples has increased in recent years. These additives are used to enhance flavor or improve food quality in the food industry. Nevertheless, many of them are very harmful to health according to scientific reports. It is quite urgent to search for an efficient approach for the degradation and detection of such a chemical to minimize its contamination in food samples. Therefore, fabrication of novel sensor for trace analysis of this type of compounds is very important in food products. Color, emulsifiers, flavoring, gelling agents, preservatives, sweeteners, anti-cake agents, antioxidants and acidulates are different categories of food additives that must be analyzed in food products. In addition, three types of electrochemical sensors can be suggested for the analysis of food additives. In the first category, the oxidation/reduction signal of food additive can be detected directly at the surface of electrode [12, 13]. In this type of sensors, the electrode is modified with conductive materials for increasing sensitivity of a sensor. In the second category, the DNA biosensors were used for analysis of food additives. Usually, the decrease in guanine and adenine base signal can be useful for the analysis of food additives. In the third category, the interaction between a conductive mediator and the food compound was suggested as an electrocatalytic interaction for analysis of food additives. In between, the electrocatalytic systems showed better selectivity for the analysis of food additives due to the selective interaction of mediator and food additive. In addition, with modification of electrode surface in the electrocatalytic systems, the sensitivity of food sensors can be improved. What are the effects of food additive? is the main question about food additive their necessary analysis. The side effects of food additive can be divided into two different categories.

1) Immediate effects such as a headache, change in energy level, alteration in mental concentration, behavior, or immune response

Long term effects such as increased risk of cancer. According to the above points about the analysis of food additives and advantage ofmodified carbon paste electrode as a voltammetric sensor, we studied the suggested sensors for food additive in continuous analysis by modified carbon paste electrodes.

Rezaei *et al*. fabricated a highly selective electrochemical platform for the analysis of rutin as a citrus flavonoid glycoside. They suggested a molecular imprinted polymer (MIP) modified carbon paste electrode as a sensor for analysis of rutin [116]. Coupling of MIP and multiwall carbon nanotubes increased the selectivity and sensitivity of sensor for the analysis of rutin. Key lock function of MIP in the carbon paste matrix improved the selectivity of the sensor for the analysis of rutin in the presence of other vitamins and biological samples. The suggested modified sensor showed a detection limit of 0.05 μM for the analysis of rutin in real samples.

Asnaashariisfahani *et al*. suggested 8,9-dihydroxy-7-methyl-12H-benzothiazolo[2,3-b]quinazolin-12-one as a mediator for electrocatalytic interaction with ascorbic acid. The mediator was incorporated into carbon paste matrix in the presence of multiwall carbon nanotubes and used as a highly sensitive sensor for ascorbic acid analysis in the concentration range 0.07-350.0 μA. The sensor showed high performance ability for analysis of ascorbic acid in the presence of acetaminophen and tryptophan. The sensor showed good ability for analysis of ascorbic acid in tablets, urine, orange, kiwi, apple, lemon and vegetable juices [117].

Pahlavan *et al*. used CdO nanoparticles and 1-butyl-3-methylimidazolium hexafluoro phosphate as two conductive mediators for the modification of carbon paste electrode. The fabricated sensor was used for the analysis of sudan I in the concentration range 0.08-550.0 μM with a detection limit of 0.05 μM. The fabricated sensor was used for the determination of sudan I in chilli sauce, chilli powder, tomato sauce and strawberry sauce [118].

Paredes *et al*. fabricated a highly sensitive D-fructose biosensor based on carbon paste electrode modified with enzyme. The fructose can be detected in the concentration range 0.2-20mM with a detection limit of 35 μM in food samples by steady-state mode. After the application of flow injection system, the fructose was detected in the range 0.5-15 M with detection limit of 115 μM [119].

Table 1. The carbon paste electrodes modified with different mediators for food compound analysis.

Mediators	LDR* (μM)	LOD* (μM)	Food Compound	Refs.
CdO nanoparticle and 1,3-dipropylimidazolium bromide	0.2-320	0.08	Curcumin	[121]
ZrO_2 nanoparticle and 1-butyl-3-methylimidazolium tetrafluoroborate	0.07-850	0.009	Vitamin C	[122]
Multi-wall carbon nanotubes and *p*-aminophenol	0.2-120	0.08	Ascorbic acid	[123]
Pt:Co nanoalloy and n-hexyl-3-methylimidazolium hexafluoro phosphate	0.1-500	0.04	Folic acid	[124]
ZnO/nanoparticles and 1-butyl-3-methylimidazolium hexafluorophosphate	0.05-550.0	0.01	Folic acid	[125]
ZnO/CNTs and 1,3-dipropylimidazolium bromide	0.02-700	0.009	Bisphenol A	[126]
NiO nanoparticle and (9,10-dihydro-9,10-ethanoanthracene-11,12-dicarboximido)-4-ethylbenzene-1,2-diol	0.01-600	0.006	Ascorbic Acid	[127]
Gold nanoparticles	0.1-2.0	0.03	Sunset yellow	[128]
nickel phthalocyanine	0.054-83.2	0.08	tert-butylhydroxyanisole	[129]
nickel phthalocyanine	4.53-226	0.771	tert-butylhydroxytoluene	[130]
poly(vinylpyrrolidone)	0.39-13.0	0.15	Rutin	[131]
cetyltrimethylammonium bromide	1.05-10.15	0.018	tert-butylhydroquinone	[132]
Unmodified	70-20000	20	Ascorbic Acid	[133]
5-amino-2′-ethyl -biphenyl-2-ol and carbon nanotube	0.2-500	0.1	Ascorbic acid	[134]
ZnO/CNTs and ruthenium(II) complex	0.008-251	0.005	Ascorbic acid	[135]
ZnO/CNTs and 1-methyl-3-butylimidazolium bromide	0.1-450	0.07	Ascorbic acid	[136]
Ethynylferrocene and carbon nanotube	0.2-250	0.07	L-cysteine	[137]
multiwall carbon nanotubes and 3,4-dihydroxycinnamic acid	0.02-140	0.009	Ascorbic acid	[138]
n-octylpyridinium hexafluorophosphate	0.5-125	0.132	Epigallocatechin Gallate	[139]
NiO/CNTs and 1-methyl-3-butylimidazolium bromide	0.08-400	0.03	Quercetin	[140]

* LOD: limit of detection; LDR: linear dynamic range

Voltammetric determination of sudan I was described by Lin *et al*. using carbon paste electrode modified with montmorillonite calcium. The sudan I analysis at the surface of the proposed sensor was performed in the concentration rage 0.2-4.03 μM with a detection limit 0.08 μM [120].

There are many published papers for the analysis of food compounds using modified carbon paste electrodes that are listed in the Table **1** [121-140]. As can be seen, different types of mediators are used for improving sensitivity and selectivity of carbon paste electrodes for food sample analysis.

4. MODIFIED CARBON PASTE ELECTRODE AS DRUG SENSOR

The analysis of drugs plays an important role in human health. There are many side effects of the consumption of drugs and especially anticancer drugs. In addition, there are several drugs for a type of illness with different side effects. The response of the body to drugs relative to the interaction of drugs with the human body is different in every individual. The determination of drugs level in the urine and blood samples helps doctor in prescribing the right drug (high level of drug in blood or low level of drug in urine confirmed good effect of drug in the human body). The fast and selective analyses of drugs in the human body are major problems in the diagnosis and treatment. Although high performance liquid chromatography is an important analytical technique for analysis of drug samples from many years, but this analytical technique isn't fast and cannot convert to portable kits. Therefore, many scientists suggested electrochemical methods as a high performance and fast analytical method for the analysis of drugs. The glucose kit is an applied example of electrochemical analytical systems. Carbon paste modified electrodes as working electrodes with simple modification process were suggested for the analysis of drugs and biological samples. Carbon paste electrode can be used di-

rectly for the analysis of drugs by recording their oxidation or reduction signals. The oxidation or reductions of drugs is relative to their structure. As example, adrenaline can be oxidised at a surface of electrode according to the below mechanism (Fig. **1**).

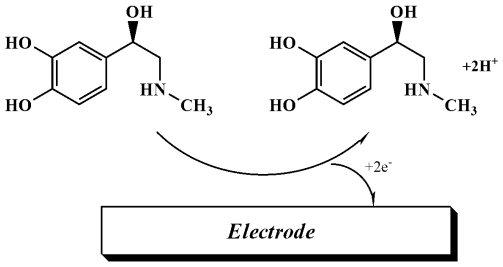

Fig. (1). The electrooxidation mechanism for adrenaline.

The moved electron from working electrode to counter electrode can be detected by an electrochemical system and recorded current is relative to the concentration of drug. The conductivity of electrode is very important for analysis of drugs in the trace level. The modification of electrode with conductive materials helps in improving the limit of detection.

Bavandpour *et al*. reported amplified carbon paste electrode with $CuFe_2O_4$ nanoparticles and 1,3-dipropylimidazolium bromide. At a biological condition, the proposed sensor showed a good ability for analysis of adrenaline in the concentration range of 0.1-400 μM. The sensor was used for the analysis of adrenaline in injection and urine samples [141].

Karimi-Maleh *et al*. suggested carbon paste electrode with ZnO nanoparticle and n-hexyl-3-methylimidazolium hexafluoro phosphate as a highly sensitive sensor for analysis of isoprenaline in the presence of aspirin. Using the proposed sensor the isoprenaline analysis was performed in the concentration range of 0.3-320 μmol L^{-1} with a detection limit of 0.09 μmol L^{-1} [142].

Ferancová *et al*. modified carbon paste electrode with β-cyclodextrin as a sensor for analysis of tricyclic antidepressants - imipramine, trimipramine and thioridazine by differential pulse voltammetric method. [143].

Gupta *et al*. suggested carbon paste electrode modified with ZnO/CNTs nanocomposite and n-hexyl-3-methylimidazolium hexafluoro phosphate as sensor for analysis of carbidopa. The conductive modifiers improved sensitivity of sensor for analysis of carbidopa and reduce overvoltage for the oxidation of this drug simultaneously. Figure (**2**) showed cyclic voltammograms of carbidopa at the surface of unmodified and modified electrodes [144].

As can be seen, the modifiers reduced the oxidation overvoltage of carbidopa and increased the sensitivity for analysis of this drug.

Shahrokhian and Asadian used thionine immobilized multi-walled carbon nanotube as a conductive mediator for modification of carbon paste electrode. The fabricated sensor was used for the analysis of ascorbic acid, acetaminophen and isoniazid [145].

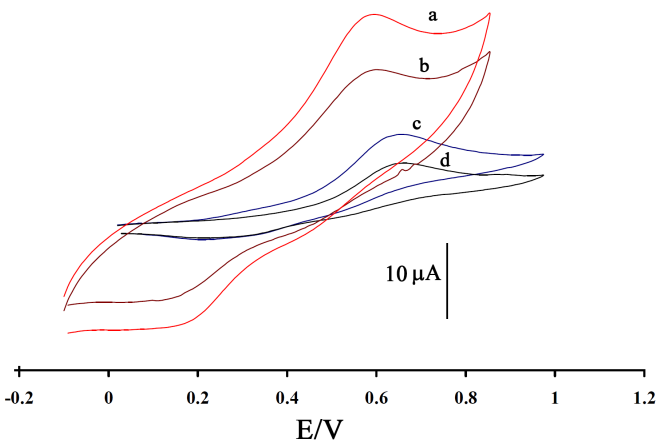

Fig. (2). Cyclic voltammograms of (**a**) carbon paste electrode modified with ZnO/CNTs and ionic liquid, (**b**) carbon paste electrode modified with ionic liquid, (**c**) carbon paste electrode modified with ZnO/CNTs and (**d**) CPE in the presence of 200 μmol L^{-1} carbidopa at pH 6.0, respectively [144].

On the other hand, the interaction between electro-active mediators with drugs can be useful for the determination of drugs. The electrocatalytic interaction with EC/ mechanism is another way for selective determination of electroactive materials. In this way, the signal of mediator before and after interaction with analyte was recorded:

$Red \rightarrow OX + ne^-$

$OX + Analyte_{(red)} \rightarrow Red + Analyte_{(ox)}$

As can be seen in the above mechanism, due to chemical reaction the oxidation signal increased and this increase can be relative to the concentration of analyte. Therefore, in this way, the mediator signal was detected and this signal can be useful for the analysis of electroactive materials. Due to the selective interaction of mediator with electroactive material, this analysis shows better selectivity for electrochemical determination of compounds as compared to direct analysis of them. Electroactive mediator can be used in two different forms for electrocatalytic analysis of electro-active materials.

1) Homogeneous form: in this form the mediator is added to the solution
2) Heterogeneous form: in this form the mediator incorporated at a surface or matrix of electrode

Due to low toxicity and better electro-catalytic interaction, the heterogeneous application of mediators suggested for an EC/ mechanism. Due to the easy modification of carbon paste electrodes with a heterogeneous mediator, this type of electrodes is recommend as working electrodes in electrocatalytic analysis. Figure (**3**) shows an example of electrocatalytic mechanism for the analysis of the drug cysteamine.

In their study, Karimi-Maleh *et al*. suggested ferrocenecarboxaldehyde (FCAD) as an electro-active mediator for the interaction and analysis of cysteamine. They used NiO/NPs as a conductive modifier for improving the conductivity of electrode surface. As can be seen in curve a, FCAD showed

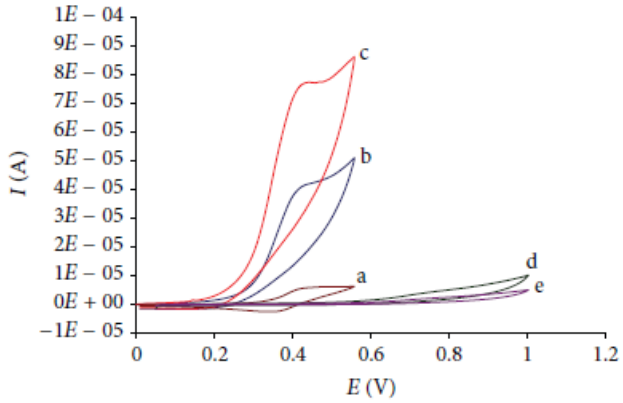

Fig. (3). Cyclic voltammograms of (**a**) the buffer solution at FCAD/NiO/NPs/CPE; (**b**) 1.0 mM cysteamine at FCAD/CPE; (**c**) 1.0 mM cysteamine at FCAD/NiO/NPs/CPE; (**d**) 1.0mM cysteamine at NiO/NPs/CPE; and (**e**) 1.0 mM cysteamine at CPE. Conditions: 0.1 mol L^{-1} PBS (pH 7.0), scan rate of 20 mVs^{-1} [164].

a quasi-reversible signal in the absence of cysteamine. After addition of cysteamine, the oxidation signal of FCAD increased and reduction signal of mediator was removed. The difference between FCAD oxidation signal before and after the addition of cysteamine can be relative to the concentration of drug.

Table **2** [146-163] presents some examples of carbon paste modified electrodes that were suggested for the analysis of drugs.

5. MODIFIED CARBON PASTE ELECTRODE AS WATER POLLUTANT SENSOR

Electrochemical methods and especially voltammetric based techniques are one of the most effective strategies for the analysis of water pollutants. There are some types of water pollutants such as toxic ions *e.g*. Pb^{2+}, As^{3+}, Cd^{2+}, *etc*., organic and inorganic molecules such as phenol, hydrazine, sulfite, sulfide, hydroxylamine, nitrite, pesticides, *etc*. ICP, atomic absorption spectroscopy (AAS), atomic emission spectroscopy (AES) are three important methods for analysis of toxic ions in water and wastewater samples. In between, ICP showed the better limit of detection for analysis of toxic ions in water samples compared to AAS and AES methods due to low detection limit and application for analysis of a wide range of ions. In addition, ICP can analyze many ions simultaneously. Nevertheless, ICP is an expensive technique and cannot be used as a portable kit for fast analysis of organic and inorganic pollutants. Therefore, electrochemical methods were suggested as a new approach for the analysis of water pollutants. From many years, dropping mercury electrode (DME) was suggested as a highly sensitive sensor for the analysis of water pollutants and especially toxic ions. As we know, the reduction window at a surface of DME is wider than other electrodes. Therefore, this type of electrode can be useful for the reduction of toxic ions and their analysis [165]. Nevertheless, oxidation window of DME is not sufficient for the analysis of organic and inorganic pollutants, DME is a toxic electrode, and application of this electrode can be harmful to the human body. Therefore, carbon paste electrodes can be a good choice for application as sensors for analysis of water pollutants with oxidation or reduction mechanism. The bubbling of nitrogen gas in the analysis sample before analysis of water pollutants based on their reduction at a surface of the modified sensor is necessary for removal of oxygen. It is well-known that the oxygen can be reduced in two steps generating large reduction peaks and removal of this compound is necessary before reduction analysis of toxic ions. Due to the advantages of carbon paste modified electrodes, many scientists recorded this type of sensors for the analysis of water pollutants. As an example, Mhammedi *et al*. reported a carbon paste electrode modified with apatite as a sensor for analysis of para-nitrophenol in river water in the concentration range 0.2-100 µM. The modified sensor shows a detection limit of 8.0 nM and used for

Table 2. The carbon paste electrode modified with different mediators for direct analysis of drugs.

Mediators	LDR (µM)	LOD (µM)	Drug Compound	Refs.
NiO nanoparticle and 1-methyl-3-butylimidazolium chloride	0.7-900	0.4	levodopa	[146]
Fe:Co Nanoalloy	0.06-600	0.03	Methyldopa	[147]
Triton X 100	0.291-62.7	0.025	acetaminophen	[148]
Triton X 100	0.291-62.7	0.084	aspirin	[148]
Triton X 100	0.291-62.7	0.083	caffeine	[148]
Unmodified	0.2-50	0.01	lansoprazole	[149]
Unmodified	0.2-50	0.025	omeprazole	[149]
Cobalt Phthalocyanine	0.4-100.0	0.14	Theophylline	[150]
Pt-Co nanoparticles and 2-(3,4-dihydroxyphenethyl)isoindoline-1,3-dione	0.07-500	0.009	N-acetylcysteine	[151]

(Table 2) contd….

Mediators	LDR (μM)	LOD (μM)	Drug Compound	Refs.
N-(4-hydroxyphenyl)-3,5-dinitrobenzamide and MgO nanoparticle	0.03-600	0.009	cysteamine	[152]
1,1′-bis(2-phenylethan-1-ol)ferrocene and ZnO nanoparticle	0.09-450	0.05	captopril	[153]
Isoproterenol and carbo nanotubes	0.3-90	0.1	captopril	[154]
cetyltrimethylammonium bromide	0.1-20	0.05	Ciprofloxacin	[155]
2-(4-oxo-3-phenyl-3,4-dihydro-quinazolinyl)-N′-phenyl-hydrazinecarbothioamide and carbon nanotubes	0.05-550	0.009	Epinephrine	[156]
N-(4-hydroxyphenyl)-3,5-dinitrobenzamide and FePt/CNTs	0.004-340	0.001	glutathione	[157]
ZnO/CNTs and 8,9-dihydroxy-7-methyl-12H-benzothiazolo[2,3-b]quinazolin-12-one	0.002-720	0.0008	glutathione	[158]
(9,10-dihydro-9,10-ethanoanthracene-11,12-dicarboximido)-4-ethylbenzene-1,2-diol and NiO/NPs	0.035-550	0.007	Captopril	[159]
1-methyl-3-butylimidazolium bromide and NiO/NPs	0.03-900	0.009	NADH	[160]
NiO/CNTs and 1-methyl-3-butylimidazolium chloride	0.05-520	0.01	morphine	[161]
ZnO/CNTs and N-(4-hydroxyphenyl)-3,5-dinitrobenzamide	0.05-800	0.01	captopril	[162]
1,3-dipropylimidazolium Bromide and NiO/NPs	0.09-750	0.05	NADH	[163]

Table 3. Carbon paste electrodes modified with different mediators for direct analysis of water pollutants.

Mediators	LDR (μM)	LOD (μM)	Water Pollutants Compound	Refs.
Ferrocene monocarboxylic acid and carbon nanotubes	0.8-700	0.42	phenylhydrazine	[170]
Ferrocene and carbon nanotubes	0.85-700	0.6	phenylhydrazine	[171]
p-aminophenol and multiwall carbon nanotubes	0.5-175	0.3	Hydrazine	[172]
dipyridyl-functionalized silica gel	0.5-100000	0.1	Silver Ion	[173]
(3,4-dihydro-4,4,6-trimethyl-2(1H)-pyrimidine thione)	0.977-76000	0.07	copper(II)	[174]
p-aminophenol and multiwall carbonnanotubes	0.2-850	0.09	sulfite	[175]
Pchloranil and carbon nanotubes	0.1-172	0.08	hydroxylamine	[176]
ZnO/CNTs and N-(4-hydroxyphenyl)-3,5-dinitrobenzamide	0.02-550.0	0.009	hydrazine	[177]
ZnO/CNTs and 8,9-dihydroxy-7-methyl-12H-benzothiazolo[2,3-b]quinazolin-12-one	0.09-350	0.04	Hydroxylamine	[178]
1,1-bis(phenylacetyl)ferrocenele and NiO/CNTs	0.5-250	0.2	Hydroxylamine	[179]

the analysis of para-nitrophenol in water samples with recovery ~86.2% [166]. Ganjali *et al*. described a carbon paste electrode modified with multiwall carbon nanotubes and nanosilica and used it for the analysis of Pb^{2+} ions in the concentration range 0.1-10000 μM. The sensor showed good ability for the analysis of Pb^{2+} ions in wastewater and black tea [167]. Ensafi *et al*. suggested carbon paste electrode modified with multiwall carbon nanotubes and ferrocenedicarboxylic acid for analysis of sulfite in the weak liquor sample with a detection limit of 0.3 μM [168].

Karimi-Maleh *et al*. used ferrocene as an electroactive mediator for the electrocatalytic determination of sulfite in the concentration range of 0.4-120.0 μM with detection limit of 0.1 μM [169]. The modified electrode was used for the analysis of the pollutant in drinking water, river water and industrial wastewater. Other examples of modified carbon paste electrodes used for analysis of different types of pollutants are listed in Table **3** [170-179].

SUMMARY AND CONCLUSION

The importance of electrochemical sensors has grown dramatically in recent years [180-184]. The working electrode is the main part of an electrochemical cell that plays an important role in the voltammetric analysis. There are many types of working electrodes for the analysis of drug, food and pollutants compounds. In between different types of working electrodes, carbon paste electrode is a great option for analysis of electroactive compounds [185-188]. Low sensitivity and high overvoltage of electroactive materials at the surface of unmodified carbon paste electrodes are major problems of this type of working electrodes. Therefore, modified carbon paste electrodes were suggested as a new type of sensor for the analysis of electroactive materials. The modified carbon paste electrodes can improve selectivity and sensitivity for the analysis of food, drug and water pollutants compounds.

CONSENT FOR PUBLICATION

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CONFLICT OF INTEREST

The authors declare no conflict of interest, financial or otherwise.

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