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Pioglitazone Hydrochloride/SDS Modified Carbon Paste Electrode for Electrochemical Determination of Dopamine: A Cyclic Voltammetric Study

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Abstract

The drug based sensor was fabricated for the electrochemical determination of dopamine, carbon paste electrode was modified using Pioglitazone Hydrochloride drug and anionic surfactant sodium dodecyl sulphate. The electrochemical parameters of the fabricated electrode were scrutinized by cyclic voltammetry and differential pulse voltammetric techniques. The modified Pioglitazone Hydrochloride/SDS carbon paste electrode had excellent electro catalytic property with promising selectivity towards the detection of dopamine in presence uric acid and ascorbic acid. The detection limit of dopamine and uric acid were found to be 1.081 nd 1.147 µM respectively. The proposed sensor was successfully fit for the analysis of dopamine in pharmaceutical sample.

Keywords: Dopamine; Uric acid; Ascorbic acid; Pioglitazone hydrochloride; Modified carbon paste electrode; Sodium dodecyl sulphate (SDS); Cyclic voltammetry

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Introduction

The modified carbon paste electrodes gaining prominence importance right now in electroanalytical techniques due to their low cost, chemical inertness, good electron transfer ability and ease of handling. The electrochemical response of carbon paste electrode mainly depends on the properties of the modifying species. The modification of the carbon paste electrode can be done by different ways like grinding in an agate mortar [1-4], electro-polymerization [5,6] and immobilization method [7]. One of the most important properties of modified carbon paste electrode is their ability to catalyze the electrode process via significant decreasing of over potential with respect to unmodified electrode. Modified electrodes are capable of enhancing the selectivity in electro analytical methods. Here we attempted to construct a new drug based sensor for the detection of neurotransmitters, drug is a substance used to diagnosis the disease. Pioglitazone Hydrochloride is an oral anti-hyperglycaemic agent; it is a class of anti-diabetic drugs are named as thiazolidinediones. It increases insulin sensitivity in target tissues. Insulin is a hormone it acts as a chemical messenger it helps our body to use the glucose to give energy. Type -2 diabetes occurs when the pancreas is not able to produce adequate insulin or the insulin does not work efficiently or the cells of the body do not respond appropriately to insulin therefore the Pioglitazone Hydrochloride is used together as monotherapy and in combination with sulfonylurea or insulin in the management of type 2 diabetes mellitus (non-insulin-dependent diabetes mellitus, NIDDM) [8,9]. Dopamine, uric acid and ascorbic acid are vital biomolecules which plays very important role in physiological process. Dopamine is one of the important neurotransmitter; it plays a very important role in cardiovascular, renal, hormonal, and central nervous and immune systems. The abnormal level of dopamine is an indicator for some diseases like Schizophrenia, Parkinson's, Alzheimer, and HIV infection [10-12]. Uric acid is the primary purine base product of metabolism in human body [13]; it is well known of its natural anti-oxidant property [14]. Uric acid produced from the breakdown of hypoxanthine by hypoxanthine-guanine phosphoribosyl transferase (HGPT) [15]. Deficiency of HGPT can affect dopaminergic neurons and dopamine concentrations [16]. The extreme abnormality of uric acid level is symptom of several

diseases such as Hyperuricemia, gout, and pneumonia [17]. Several methods have been proposed for simultaneous determination of dopamine in presence of uric acid and ascorbic acid, such as capillary electrophoresis [18] chromatography [19,20], fluorescence [21]. However electrochemical techniques are more attractive and fascinating due to their low cost and less time consumption, but mainly they are suffering from selectivity because dopamine, uric acid, ascorbic acid have close oxidation potentials so it's very complicated to identify individual peaks in voltammogram. Electroactive nature of dopamine, uric acid and ascorbic acid makes their detection possible by electrochemical methods based on anodic oxidation [22,23].

Surfactants are surface active agents; the surfactants are referred to as amphiphiles. The polar hydrophilic part of the molecule is called lipophobic group and the non-polar hydrophobic part is called lipophilic group, the hydrophilic part of the molecule is called head and hydrophobic part is called tail. Sodium dodecyl sulphate is a class of anionic surfactant. The surfactants have numerous applications in electrochemistry like electroplating [24], corrosion [25], fuel cells [26], electro catalysis [27], and electro analysis [28]. The electron transfer process for the oxidation of dopamine in presence of anionic surfactant Sodium dodecyl sulphate is quicker due to electrostatic attraction of positively charged dopamine with anionic surfactant Sodium dodecyl sulphate [29-32].

In this present work, we introduced a Pioglitazone Hydrochloride/SDS modified carbon paste electrode for the detection of dopamine. The fabricated sensor selectivity was examined by selective determination of dopamine in presence of uric acid and ascorbic acid. In order to evaluate the drug based sensor capability the determination of dopamine in pharmaceutical preparations were performed. The detection limit observed for dopamine and uric acid was compared with several other modified electrodes in **Table 1**. The structure of Pioglitazone Hydrochloride as shown in **Scheme 1**.

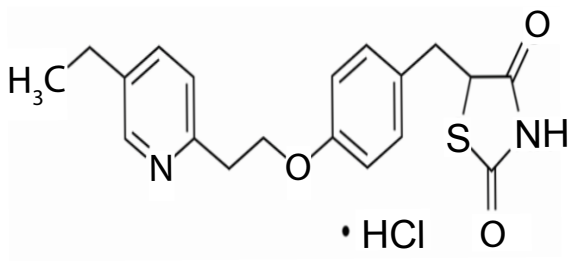
Experimental Part

Reagents and chemicals

Pioglitazone Hydrochloride was received from Biocon limited Bangalore. Dopamine and silicon oil were purchased from HI media. Graphite powder was purchased from Lobe chemical company with analytical grade used without further purification. Sodium dihydrogen phosphate monohydrate and disodium hydrogen phosphate were purchased from Merck. Dopamine stock solution was prepared in 0.1 M perchloric acid, Sodium dodecyl sulphate, potassium ferro cyanide and KCl was prepared in double distilled water.

Table 1 Detection of DA in injection samples (n=3).

Sample	DA added (M)	Found (M)	Recovery (%)
1	0.1×10^{-4}	0.9985×10^{-4}	99.85
2	0.2×10^{-4}	0.1988×10^{-4}	99.42
3	0.3×10^{-4}	0.2929×10^{-4}	97.63



Scheme 1 The structure of Pioglitazone Hydrochloride.

Apparatus

Cyclic voltammetric experiments were performed on a model CHI-660c (CH Instrument-660 electrochemical work station). The entire experiments were carried out in a conventional three electrode cell. The electrode system includes a bare carbon paste electrode (BCPE) Pioglitazone hydrochloride/SDS MCPE as working electrode, platinum wire and saturated calomel as a counter and reference electrode.

Electrode preparation procedure

Preparation of bare carbon paste electrode: The bare carbon paste electrode was prepared by manual grinding of graphite powder and silicon oil at a ratio of 70:30 (w/w) in an agate mortar to get homogenous paste. The prepared carbon paste was firmly packed into the cavity of a homemade electrode and then polished the surface by rubbing on a weighing paper.

Modification of bare carbon paste electrode: The modified Pioglitazone Hydrochloride carbon paste electrode was prepared by using 5 mg of Pioglitazone Hydrochloride, graphite powder and silicon oil at the ratio of 70:30 (w/w) in an agate mortar by manual grinding to get homogenous paste. The obtained paste was tightly packed into the cavity of a homemade electrode, polished the surface by rubbing on a weighing paper then immobilize the SDS solution on the surface of Pioglitazone hydrochloride modified electrode to fabricate Pioglitazone hydrochloride/SDS electrode. Later the electrode was systematically rinsed using distilled water to remove the unabsorbed modifier and dried in air at room temperature.

Results and Discussion

The electrochemical response of potassium ferrocyanide at pioglitazone hydrochloride/SDS modified carbon paste electrode

The electrochemical characterization of Pioglitazone Hydrochloride/SDS modified carbon paste electrode was carried out by using potassium ferrocyanide and KCl solution as a supporting electrolyte to check the current improvement property of the fabricated electrode. **Figure 1** shows the cyclic voltammograms obtained for the electrochemical response of potassium ferrocyanide at bare carbon paste electrode (dotted line), Pioglitazone Hydrochloride modified carbon paste electrode (solid line), Pioglitazone Hydrochloride/SDS modified carbon paste electrode (dashed line) for 1mM potassium ferrocyanide in 1M KCl solution with scan rate of 100 mV/s. The Pioglitazone

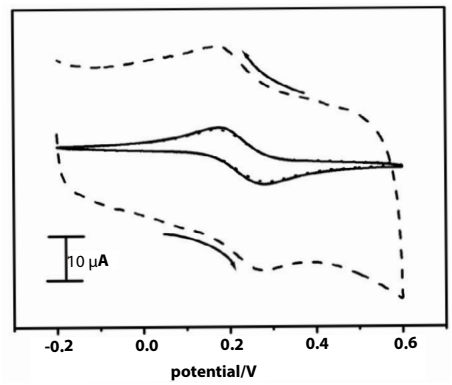


Figure 1 Cyclic voltammograms of 1 mM $[K_4Fe(CN)_6]$ at BCPE (dotted line), Pioglitazone Hydrochloride/MCPE (solid line) and Pioglitazone Hydrochloride/SDS MCPE (dashed line) in 1 M KCl scan rate 100 mV/s.

Hydrochloride modified carbon paste electrode shows small increased current signal as compared to the bare carbon paste electrode, However the Pioglitazone Hydrochloride/SDS modified carbon paste electrode exhibit a well defined redox peak with great enhancement of peak current as compared to the bare carbon paste electrode. It reveals the excellent sensitivity of Pioglitazone Hydrochloride/SDS modified carbon electrode towards the electrochemical detection of dopamine.

Electrochemical response of dopamine at pioglitazone hydrochloride/SDS modified carbon paste electrode

The **Figure 2** shows cyclic voltammograms of 5 µM dopamine at bare carbon paste electrode (dotted line), Pioglitazone hydrochloride modified carbon paste electrode (solid line), Pioglitazone Hydrochloride/SDS modified carbon paste electrode (dashed line) in 0.2 M phosphate buffer solution at pH 7.2 with a scan rate of 100 mV/s. This result favour that the electrochemical response of dopamine at Pioglitazone Hydrochloride/SDS modified carbon paste electrode was increased remarkable as compared to the bare carbon paste electrode and Pioglitazone Hydrochloride modified carbon paste electrode. It specifies that the electrochemical response of dopamine was enhanced at Pioglitazone Hydrochloride/SDS modified carbon paste electrode, due to enhanced accumulation of protonated dopamine via electrostatic interaction with negatively charged Sodium dodecyl sulphate at modified electrode.

Effect of scan rate at Pioglitazone hydrochloride/SDS modified carbon paste electrode

The different scan rate study was performed to understand the electrochemical mechanism of Pioglitazone Hydrochloride/SDS modified electrode, it displays a reflective effect on the oxidation peak current of dopamine. The **Figure 3a** shows cyclic voltammograms for 5 µM dopamine at Pioglitazone Hydrochloride/SDS modified carbon paste electrode under varying scan rate from 50 to 550 mV/s, at pH 7.2 in 0.2 M phosphate buffer solution as a supporting electrolyte. The scan rate altering studies shown that the current enhanced linearly with increasing scan rate from 50 to 550 mV/s. The anodic peak potential shifted towards positive direction and cathodic peak potential shifted towards negative direction with increase in scan rate. In **Figure 3** the graph of anodic peak current (Ipa) versus square root of scan rate gave a straight line with correlation coefficient value of 0.9960, it indicates that the electron transfer reaction was under diffusion controlled [33-37].

Effect of different concentration of dopamine at pioglitazone hydrochloride/SDS modified carbon paste electrode

The **Figure 4a** shows the cyclic voltammograms for different concentration of dopamine from 10 to 50 µM in 0.2 M phosphate buffer solution of pH 7.2 with a scan rate of 100 mV/s at Pioglitazone Hydrochloride/SDS modified carbon paste electrode. The different concentration of dopamine was prepared to investigate its redox behaviour; the redox peak current was steadily increased with increasing the concentration of dopamine and slightly the anodic peak potential shifted towards positive direction, cathodic peak potential shifted towards negative direction. The **Figure 4b** shows the graph of Ipa versus concentration of dopamine and it results that the anodic peak current was directly proportional to the concentration of dopamine.

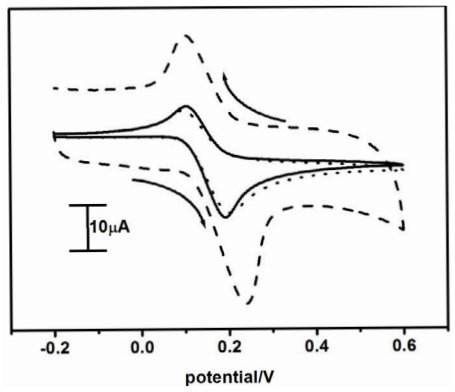


Figure 2 Cyclic voltammograms of 5 µM dopamine in 0.2 M phosphate buffer solution, pH 7.2 with scan rate of 100 mV/s at BCPE (dotted line), Pioglitazone Hydrochloride/MCPE (solid line) and Pioglitazone Hydrochloride/SDS MCPE (dashed line).

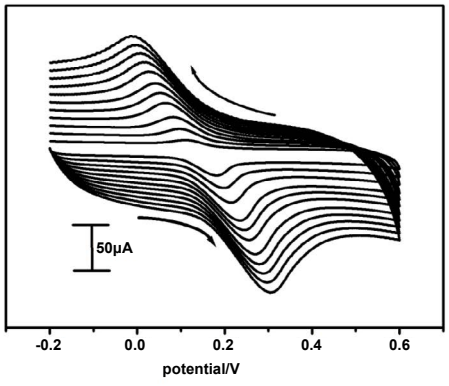


Figure 3a Cyclic voltammograms of dopamine at different scan rate (50 to 550 mV/s).

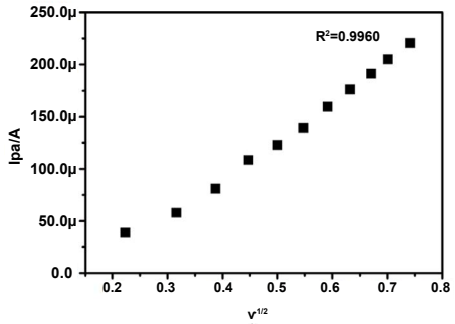


Figure 3b Graph of current versus square root of scan rate.

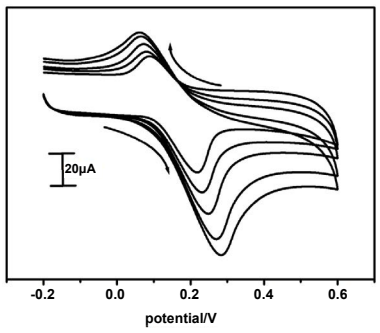


Figure 4a Cyclic voltammograms of different concentration of dopamine at Pioglitazone Hydrochloride/SDS MCPE in 0.2 M PBS at pH 7.2.

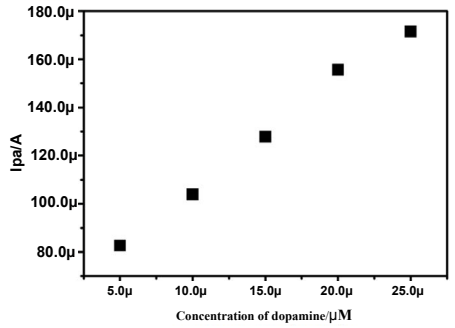


Figure 4b Graph of current versus different concentration of dopamine at scan rate of 100 mV/s at pH 7.2.

Effect of SDS concentration and immobilization time

The **Figure 5a** shows the graph of anodic peak current versus different concentration of SDS at Pioglitazone Hydrochloride/SDS modified carbon paste electrode. The result reveals the influence of SDS to enhance the electrochemical property of modified Pioglitazone Hydrochloride carbon paste electrode. It helps to optimise the condition of modified carbon paste electrode to get maximum peak current. Varying concentration of SDS on the surface of Pioglitazone Hydrochloride modified carbon paste electrode results increase in anodic peak current from 1 to 5 µl, further increasing the SDS concentration the anodic peak current goes on decreases from 5 to 25 µl. In the same way, the immobilization time also influence the anodic peak current of dopamine, it was examined by changing the immobilization time on the surface of Pioglitazone Hydrochloride modified carbon paste electrode. The **Figure 5b** shows the graph anodic peak current (Ipa) versus immobilization time it specifies that the anodic peak current achieved more at 5 min after that anodic peak current decreases. Therefore, for further electrochemical studies we selected 5 µl of SDS with immobilization time 5 min to fabricate Pioglitazone Hydrochloride carbon paste electrode.

Effect of pH for electro-oxidation of dopamine at pioglitazone hydrochloride/SDS modified carbon paste electrode

The effect of supporting electrolyte pH for the electro-oxidation of dopamine was investigated by cyclic voltammetry to realize the mechanism of electrochemical reaction of dopamine at Pioglitazone Hydrochloride/SDS modified carbon paste electrode. The results in **Figure 6a** reveal that the pH value of supporting electrolyte influences the peak current and peak potential value of analyte. The electro-oxidation of dopamine was studied with increase in the pH value from 6.0 to 8.0 the anodic peak potential shifted negatively from 313 to 172 mV at a scan rate of 100 mV/s. The result confirms that the oxidation peak potential is pH dependent, it indicates that protonation/deprotonation is taking part in electrochemical process. The potential diagram **Figure 6b** was constructed by plotting the graph of anodic peak potential verses pH of supporting electrolyte, the slope found to be 60 mV/pH unit, it indicates that the equal number of protons and electrons were involved in electrochemical oxidation

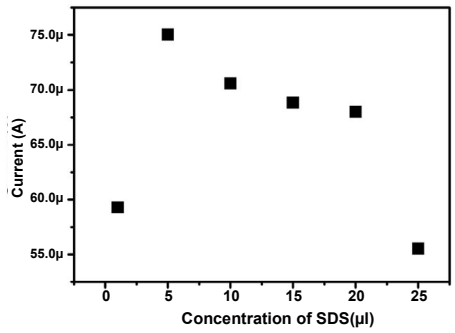


Figure 5a Graph of current versus different concentration of SDS at Pioglitazone Hydrochloride MCPE in 0.2M PBS of pH 7.2.

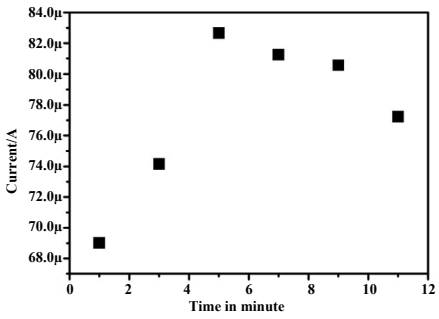


Figure 5b Graph of current versus immobilization time.

of dopamine [38]. The anodic oxidation peak current (Ipa) of dopamine increases gradually with increasing pH from 6.0 to 7.0 then peak current decreased and the maximum current achieved at pH 7.0 as shown in **Figure 6c** hence the physiological pH 7.2 was chosen to carry out for extended work.

Simultaneous detection of dopamine, uric acid and ascorbic acid

The main aim of this method was to detect dopamine, uric acid and ascorbic acid in a mixture because they are coexisting in physiological sample. The **Figure 7** shows the cyclic voltammograms obtained for the electrochemical response 5 µM DA, 10 µM UA and 10 µM AA in 0.2 M phosphate buffer solution of pH 7.2 at BCPE (solid line) Pioglitazone Hydrochloride/SDS MCPE (dashed line). The bare carbon paste electrode resulting overlapped peak and it fails to locate individual anodic peak potential for dopamine, uric acid, and ascorbic acid, though the Pioglitazone Hydrochloride/SDS modified electrode evidently shows three well distinct peaks at oxidation peak potentials 163 mV, 287 mV and -50 mV for dopamine, uric acid, and ascorbic acid with great improvement of anodic peak current. The observed result reveals that the selectivity of the modified carbon paste electrode towards the detection of dopamine in presence of uric acid and ascorbic acid.

Interference study

The interference study was carried out by differential pulse voltammetry; it offers better current sensitivity with peak resolution. The **Figure 8a** shows differential pulse voltammograms for mixture solution of dopamine, uric acid and ascorbic acid, it showed well resolute anodic peaks at peak potential value for 8 µM dopamine at 112 mV, 12 µM uric acid at 242 mV and 10 µM ascorbic acid at -62 mV. The simultaneous determination of dopamine, uric acid and ascorbic acid in a mixture was investigated by changing the concentration of one species and concentration of other two species kept constant. The **Figure 8b** shows that the anodic peak current of dopamine was increased with respect to their concentration from 3 to 18 µM while the concentration of uric acid 5 µM and ascorbic acid 8 µM was kept constant. The **Figure 8c** shows differential pulse voltammograms of different concentrations of uric acid with fixed concentration of dopamine 3 µM and ascorbic acid 8 µM. The concentration of uric acid was increased from 5 to 30 µM and it results enhanced anodic peak current was directly proportional to the concentration of uric acid. The result authenticates that anodic peak current and concentration of species have a linear relationship. Varying the concentration of one analyte does not alter the peak current and the peak potential of another analyte. The limit of detection was calculated by using equation (2) [39]

$$LOD=3S/M \quad (2)$$

Where S is the standard deviation and M is the slope obtained from the calibration plot. The detection limit of dopamine and

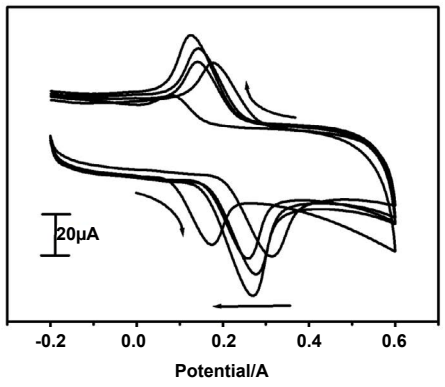


Figure 6a Cyclic voltammograms of 10 µM DA for different pH (from 6 to 8 pH) at Pioglitazone Hydrochloride/SDS MCPE.

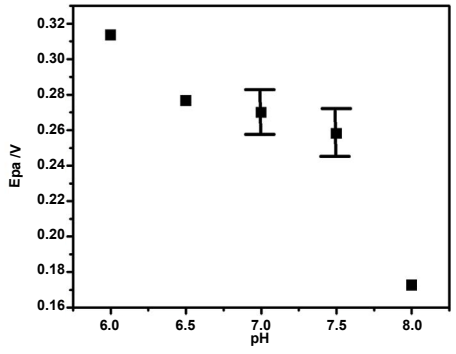


Figure 6b Graph of Epa versus pH.

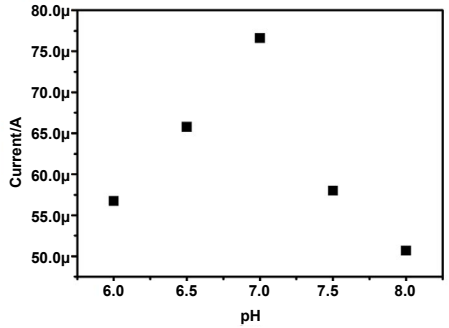


Figure 6c Graph of Ipa versus pH.

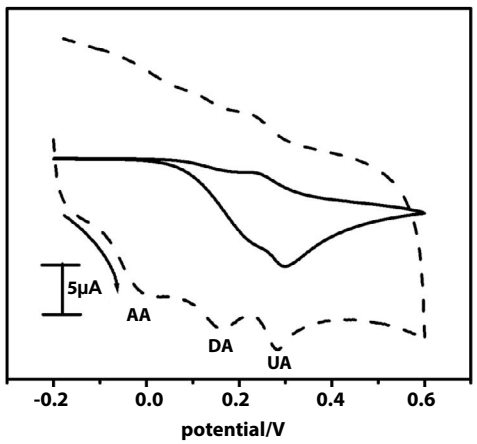


Figure 7 Cyclic voltammograms of 5 µM dopamine, 10 µM uric acid and 10 µM ascorbic acid in 0.2 M phosphate buffer solution of pH 7.2 at BCPE (solid line) and Pioglitazone Hydrochloride/SDS MCPE (dashed line) with scan rate of 100 mV/s.

Table 2 Detection limit for dopamine previous and present work.

Analyte	Electrode	Method	Detection limit (µM)	References
DA	Ag/Ag_2S-CNT-Nafion	DPV	4.7	[40]
DA	SWCNT/GCE	DPV	7.0	[41]
DA	Fc-MCPE	CV	9.4	[42]
DA	Banana/MWCNTs/MCPE	DPV	2.0	[43]
DA	Pioglitazone Hydrochloride/SDS MCPE	DPV	1.08	Present work
UA	α-CD/CNT/CPE	DPV	5.0	[44]
UA	CNT/PEDOT	DPV	10.0	[45]
UA	Pioglitazone Hydrochloride/SDS MCPE	DPV	1.14	Present work

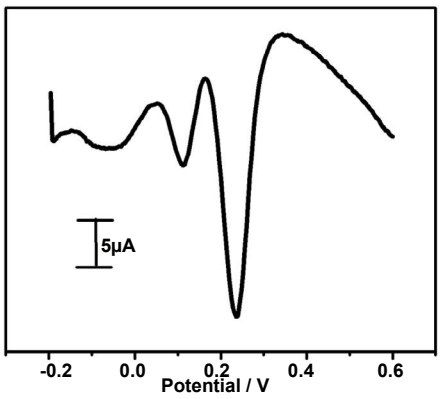


Figure 8a Differential pulse voltammogram of 8 µM dopamine, 10 µM ascorbic acid and 12 µM uric acid in 0.2 M phosphate buffer solution of pH 7.2 at Pioglitazone Hydrochloride/SDS MCPE.

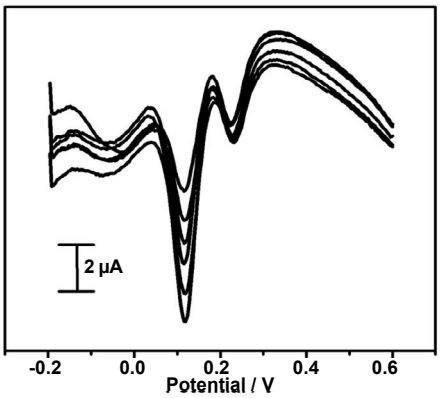


Figure 8b Differential pulse voltammograms of (a) 3 µM, (b) 6 µM, (c) 9 µM, (d) 12µM, (e) 15 µM, (f) 18 µM dopamine in 0.2 M phosphate buffer solution of pH 7.2 in presence of 5 µM uric acid and 8 µM ascorbic acid at Pioglitazone Hydrochloride/SDS modified carbon paste electrode.

uric acid were found to be 1.081 and 1.147 µM respectively. The results authenticate the prospect of the simultaneous detection of dopamine in presence of uric acid and ascorbic acid at Pioglitazone Hydrochloride/SDS electrode. These studies powerfully recommend that the Pioglitazone Hydrochloride/SDS modified carbon paste electrode may be used in electrochemical sensor applications.

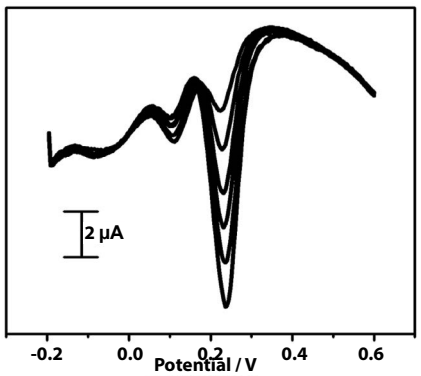


Figure 8c Differential pulse voltammograms of (a) 5 µM, (b) 10 µM, (c) 15 µM, (d) 20 µM, (e) 25 µM, (f) 30 µM uric acid in 0.2 M phosphate buffer solution of pH 7.2 in presence of 3 µM dopamine and 8 µM ascorbic acid at Pioglitazone Hydrochloride/SDS modified carbon paste electrode.

Real sample analysis

To evaluate the applicability; the constructed sensor Pioglitazone Hydrochloride/SDS modified carbon paste electrode was used for the analysis of dopamine in injection sample. The dopamine injection was purchased from Sterile Specialities India Pvt. Ltd with a specified content of dopamine of 40 mg/mL; the dopamine injection sample was diluted with 0.2 M phosphate buffer solution for suitable concentration. The results shown in **Table 2** indicates that the projected drug based modified carbon paste electrode retained its efficiency for the determination of dopamine in injection sample with recovery in the range from 97.63 -99.85%.

Conclusion

The present work depicts the construction of drug/SDS based sensor and its application for the electrochemical analysis of dopamine. The differential pulse voltammetric response has shown promising for the selective detection of dopamine without interference at modified electrode. The modified Pioglitazone Hydrochloride/SDS electrode had superior characteristics such as sensitivity, selectivity with low detection limit; the projected sensor is fit for real sample analysis of dopamine in injection sample. The significant characteristics of the drug based modified carbon paste electrode will expand its application in electrochemical field for the determination of other drugs and neurotransmitter.

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