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Full Paper

Thiosemicarbazide Poly(paraphenylenediamine) Fastened on Modified Carbon Paste Electrode Advancing Voltammetric Determination of Dopamine and Uric Acid

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Abstract- A film is developed on the surface of the carbon paste electrode by electro-ionic depositing of thiosemicarbazide (TSC) chemical substance and electropolymerized with paraphenylenediamine (PPD) and the modified carbon paste electrode (MCPE) was constructed at physiological pH 7.5 in PBS buffer solution. The bioactive molecules of dopamine (DP) and uric acid (UA) electro-chemical distinctiveness were studied by cyclic voltammetry method. TSC/poly(paraphenylenediamine) MCPE was used to examine DP and UA by cyclic voltammetric (CV), and differential pulse voltammetric (DPV) techniques. The addition of research constraints distressing the electrocatalytic action on MCPE such as the effect of pH, scan rate, and concentration differentiations are studied. The limit of detection of DP and UA were premeditated by CV as 1.34 µM and 4.19 µM, respectively. The modified electrode illustrated advanced sensitivity and selectivity for the synchronized determination of DP and UA, and the unclean consequence is also eradicated by this MCPE.

Keywords- Dopamine; Uric acid; Thiosemi carbazide; Paraphenylene diamine; Carbon paste electrode; Modified carbon paste electrode; Differential pulse voltammetry

1. INTRODUCTION

The improvement of voltammetric detectors meant for DA and UA has penetrated considerable notice right through the past couple of eras. Dopamine (DA) is solitary of the manifestly departing on catecholamine it executes a decisive utility as a neurotransmitter in the sports of the central and peripheral nervous systems [1-3]. Excessive irregularities of DA extent are signs of abundant diseases, besides Schizophrenia and Parkinsonism, patients under attack by this disarray put forward a low level of DA. So, the detection of DA-contained samples executes a decisive component inside the come across at cadaver arrangement mechanism and systematic diagnoses [4,5]. Particularly the perfection of voltammetric sensors for the determination of neurotransmitters, general dopamine (DA), and diverse catecholamines has attained a diversity of awareness. Amongst the household of catecholamine, DA acquired heaps curiosity, owing to the reality that the extrude in DA row proffers a dirt-free proceeding relating to the proficiency of intellect functions, broad of in advance acquaintance of and nostalgia creation, the pathological and physiological procedure of Parkinson's disease [6]. As a conclusion effect of this, catecholamine remedies at the minute are lengthily exploited in assorted remedial fields such as medication for hypertension, bronchial asthma, Parkinson's disease, cardiac surgical conduct myocardial infarction, etc. There are a range of approaches have been planned for the quantitative and qualitative drives of DA within the diagnosis [7,8].

The significance of UA within the body is well-identified and proven. UA (trihydroxy purine) is the primary product of purine metabolism [9,10]. Physiological UA serum situations range from 41 to 88 mg mL^{-1} and urinary excretion is generally 250–750 mg per day. UA is an item for consumption of metabolic goes kaput of purine nucleotides and the abnormal position of UA results in several conditions like gout, Hyperuricemia, and pneumonia. XA is an imperative midway of purine metabolism and the idiosyncrasy of XA shows the ways to xanthinuria and its remedial position is 10–20 mg mL^{-1} in blood [11]. Its abnormal attention position in a mortal body may be the warning sign of several circumstances, comparable as gout, Hyperuricemia, Lesch-Nyhan pattern, Leukemia, pneumonia, and so on are also associated with enhanced urate situations. So, it's advantageous to have a simple and direct system for casing the consideration of UA in usual fluids [12-15]. Recently many methods namely chemiluminescence [16]. High-performance liquid chromatography [17]. Electrochemical methods are widely applicable in identifying the uric acid properties [18-20]. Spectrophotometric [21,22].

Thiosemicarbazide (TSC) is a chemical compound by having the molecular formula $H_2NC(S)NHNH_2$ with the insertion of the basic nature of NH (amine) was interior. They are normally used as strong legends for transition quintessence. In this thiosemicarbazide, so many types of Hs of the parent organic molecules are there in the form of substituents [23-24]. In thiosemicarbazide the bond length between C=S is 1.63 A^0, in thioformaldehyde also consisting the same bond lengths due to steric relations. So, in thiosemicarbazide by multiple bonds and on their terminals presence of amine groups is cohesive with carbon paste and acts as a good catalytic nature which is shown in this research experiment. Unchecked dialkylthiones polymerize or oligomerize but thiocamphor is a well-characterized red solid [25,26].

Paraphenylenediamine (PMDA) is an organic emulsion composite with the molecular formula $C_6H_4(NH_2)_2$. This outer growth of aromatic amine is a colorless solid; when this composite turns blackened appropriate to distinctive air oxidation. This is significantly worn as a molecular atom of engineering polymers and mixes in the vein of Kevlar. It is as well a module in hair coloring and is sometimes used as a cover for henna [27]. Paraphenylenediamine (PMDA) has a temperament to copolymerize on carbon paste electrodes by the being there of aniline functional was electrochemically performed by cyclic voltammetry analysis 0.2M PBS solutions. The work of art examination of uric acid by way of carbon paste electrode with thiosemicarbazide mixture PMDA cross–linkage and branched reaction process during electropolymerization on carbon paste electrode [28]. A covering free of charge awake in stream bio-catalyzed electrolysis reactor united with an aerobic make contact through oxidation reactor was industrialized. Alizarin unheroic reactor as the mode of azo coloring was competently rehabilitated to PMDA and 5,5-aminosalicylic acid [29].

Dopamine (DA) is solitary of the manifestly departing on catecholamine it executes a decisive utility as a neurotransmitter in the sports of the central and peripheral nervous systems, The significance of UA within the body is well identified and proved. UA (trihydroxypurine) is the primary item for the consumption of purine metabolism. In the nearby exertion thiosemicarbazide-poly(paraphenylenediamine) was worn as a modifier for the fabrication of carbon paste electrode (MCPE), which was shortly old for the voltammetric fortitude of DA and CC in a binary assortment by cyclic voltammetric (CV) and Differential Pulse Voltammetric (DPV) techniques. The fictitious carbon paste electrode not merely exposed electrocatalytic movement in the analysis; in toting up it estranged the voltammetric signals of the DA and UA in a double mixture at physiological pH. For that reason, the fictitious electrode containers subsist worn as a potential sensor in the analysis of bioactive analytes of DA and UA.

2. EXPERIMENTAL SECTION

2.1. Chemicals

Dopamine hydrochloride, uric acid, thiosemicarbazide, and graphite powder were purchased from Sigma Aldrich, India. The solid-state of paraphenylenediamine (PMDA) was prepared in stock solution form 25×10^{-3} M by dissolving into distilled water. Uric acid and mono sodium dodecyl sulphate ($NaH_2PO_4.H_2O$) and di sodium dodecyl sulphate ($Na_2H_2PO_4.H_2O$) chemicals and other chemical compounds utilized in the studies was of analytical grade and were used without purification.

2.2. Equipment

A three–three-electrochemical arrangement together with a bare electrode or a modified glassy carbon electrode (MGCP) or a carbon paste electrode, as working electrodes, Ag/AgCl electrode as reference electrode, and a platinum electrode as an auxiliary electrode were used. The electrochemical measurements were conducted using CHI-660c (CH Instrument-660 electrochemical workstation) connected to a computer with a general purpose electrochemical system software package (NOVA). At room temperature 25±0.5°C all experiments were presented.

2.3. Preparation of bare carbon paste electrode

During the progress of bare carbon paste electrode (BCPE), a homogenous paste of carbon was fashioned by appropriate coming together of 70% Graphite and 30% silicon oil in an agate mortar, then the twisted carbon pastes were initiated contained by a hollow space of Teflon tube and soften on a weighing paper. Now the electrode was made available by way of an electrical drop a line to by copper wire which was connected to the paste at the end of the tube. By this electrode, the acquired voltammogram of bioactive analytes is shown in Figure 1.

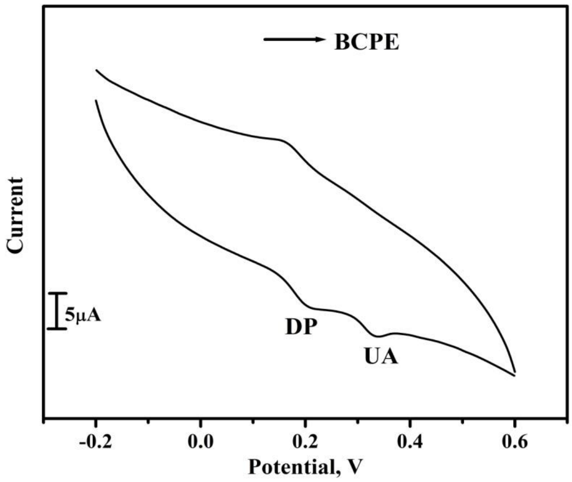


Figure 1. A cyclic voltammogram bare carbon paste electrode of 0.2 M PBS at pH7.5

3. RESULTS AND DISCUSSION

3.1. TSC electrochemically deposited and paraphenylenediamine electropolymerization on the exterior of CPE and its temperament

A carbon paste electrode gets changed and distorted into fictional throughout TSC which is later ionically deposited with the electropolymerisation of 0.1 mM paraphenylenediamine through cyclic voltammetry in 0.2 M PBS at physiological pH of 7.4 in a potential range from –0.6 V to +1.6 V and scan rate of 0.05 Vs^{-1}. The misrepresentation was positioned contained by a potential range a great deal fewer than +0.6 V [29-32]. If the polymer breadth of the motion portrait prominent it discontinues the approach of changing of electrons inside the 0.05 Vs^{-1} test price. The cyclic voltammetry showed that the anodic peak current (become visible at 0.22 V for DA and 0.32 V for UA the oxidation of PPA monomer on CPE) regularly declined among cyclic time. The rising in redox current value here CVs be shown in Figure 2, which is beginning signals of redox potential values of DA and UA moreover imitating in the progress of electro active layer on plane of CPE.

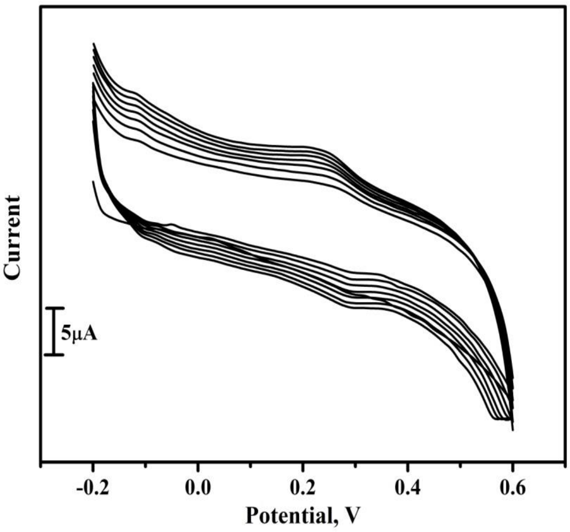
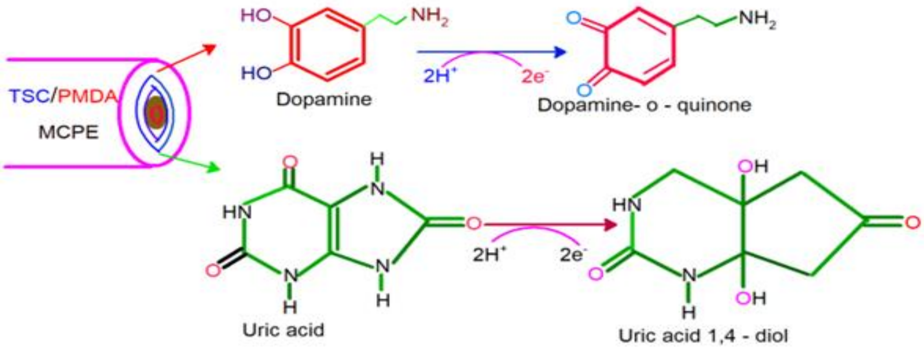


Figure 2. Cyclic voltammograms obtained for the electro-ionic deposit of 1.0 mM TSC, and electropolymerisation of PMDA on the surface of CPE; 1.0 mM of PMDA solution in 0.2 M PBS of pH 7.5 with the scan rate of 0.1 Vs^{-1} for 15 cycles

Then, getting higher in the redox peak current be invariable and steadier behind a number of uncertain sweepings up representing the extent of diffusion in electro active growth of polymer [31,32]. Whilst the sweep sectors be greater than before the fictitious electrode demonstrates superior electrocatalytic reply, while here exist further than 15 polymerizing cycles of the peak currents in the strength of mind of DA and UA start in on steadier. During adding up the elevated the compactness (solidity) of the polymeric film, the functioning electrode analogous electro- catalytic activity fades away. As a result, in producing a stable TSC and poly (paraphenylenediamine) MCPE for the resolution fifteen cycles was in use while finest [30,31]. The probable electro ionic authentications of the electro polymerization of paraphenylenediamine at the CPE facade are revealed in Scheme 1.



Scheme 1. The electro ionic deposition and electropolymerisation of the carbon paste electrode surface and dopamine and uric acid oxidation reaction in electrochemical cell

3.2. Explanation of TSC/ poly (paraphenylenediamine) MCPE

Figure 3 explains it without a doubt and gives an idea about the different voltammograms of TSC/Poly(paraphenylenediamine) modified carbon paste electrode, cyclic voltammogram of BCPE (dashed line), cyclic voltammogram of MCPE with TSC (solid line), and the cyclic voltammogram of TSC/poly(paraphenylenediamine) MCPE (dotted line). Cyclic voltammetry peaks were recorded at the scan rate of 0.05 Vs^{-1} at a supporting electrolyte of 1 M KCl.

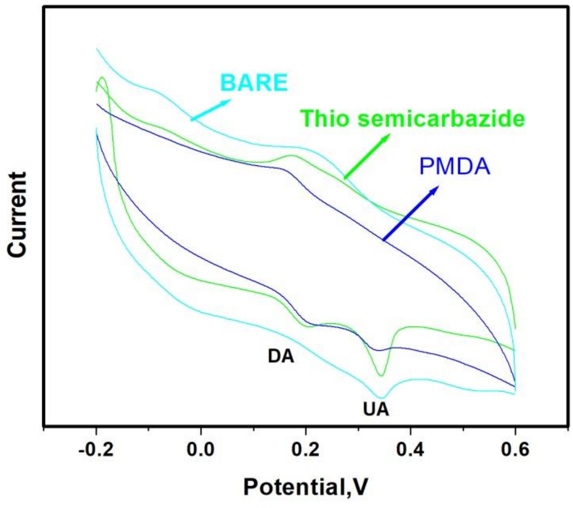


Figure 3. Cyclic voltammograms for simultaneous determination of a binary mixture of DP and UA (0.1 mM) at BCPE (dashed line) and TSC and PMDA MGCE (solid line) at a scan rate of 0.05 Vs^{-1}

The obtained voltammograms of the modified carbon paste electrode among TSC and poly(paraphenylenediamine) point to remarkable updating in the electron transferring process. The advanced result of electron movement at the voltammetric response was obtained from supporting electrolytes at MCPE with TSC and poly(paraphenylenediamine) confirming the morphology was satisfactorily customized and the result shows the electrocatalytic activity.

The peak current can be calculated by using the Randles–Sevcik equation (1) [6,31,32]:

$$I_p = 2.69\times10^5 n^{3/2} A D^{1/2} C_o v^{1/2} \qquad (1)$$

where the concentration of the electroactive species is C_o in (mol cm^{-3}), the peak current in I_p is measured in A, the transferred number of electrons is n, the diffusion-coefficient is D, the scan rate υ in (Vs^{-1}), and A is the electroactive surface area (cm^2). For TSC/poly (Paraphenylenediamine) MCPE, the surface area of electroactive is maximum (0.0435 cm^2) as compared with bare CPE (0.027 cm^2).

In this anticipated system, the oxidation stair of uric acid is transformed to uric acid 1,4 diol. In this oxidation progression, two numbers of electrons are involved. It was shown in Scheme 1.

3.3. Optimization of faltering conditions

3.3.1. The effect of Scan rate

The scan rate effect of both analytes was examined in this parameter for this 0.5 mM DA and 0.5 mM UA by CV technique at 0.2 M PBS at TSC/poly(paraphenylenediamine) (Figure 4, 5, 6). The electrochemically modified carbon paste electrode shows very good and particularly noticeable enhancing redox peak current with broadened of the purposeful scan rates, and an incredibly slightest inconsistency of redox peak potentials container subsist conscious. This was done by applying the Randles–Sevcik equation. During the order of the proven process of the electrode, the graph plotted alongside current (I_{pa}) *vs.* square root of scan rates ($v^{1/2}$). The graph is a straight line for DP and UA respectively (Figure 7 and 8).

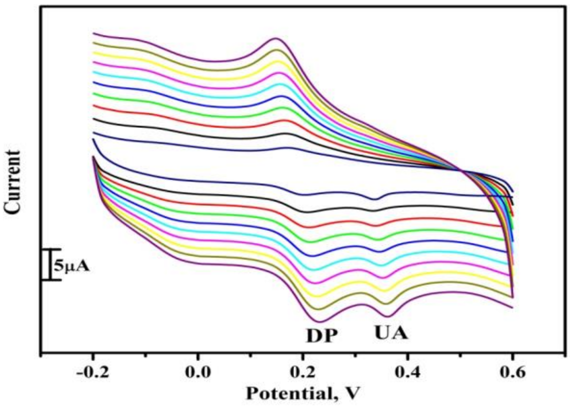


Figure 4. Cyclic voltammograms of 0.1 mM DP and 0.1 mM UA in 0.2 M PBS solution of pH 7.4 at TSC and poly(paraphenylenediamine) MGCE with different scan rates (0.05-0.5 Vs^{-1})

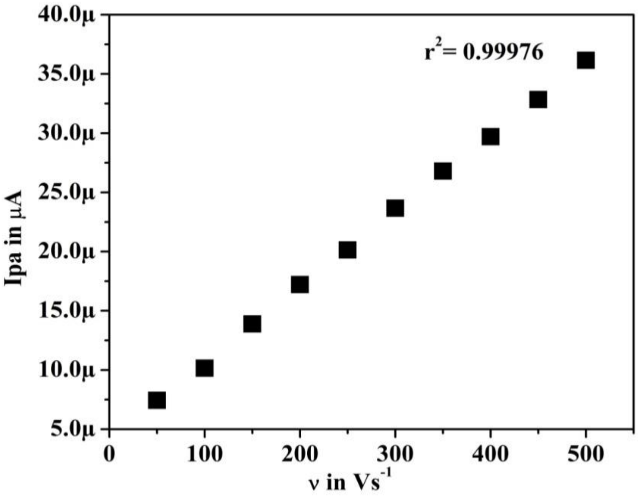


Figure 5. Graph of anodic peak current of UA versus scan rate

The graphs achieved were just about straight lines of anodic peak currents that were linearly compared to the scan rate and square root of scan rates and their correlation. Coefficients for I_{pa} *vs.* $\nu^{1/2}$ are $r^2 = 0.99428$, and $r^2 = 0.99976$ for DP and UA and I_{pa} *vs.* scan rate (ν) is $r^2 = 0.99976$, and $r^2 = 0.99917$ for UA and DA contained by the assortment of 0.002–0.2 Vs^{-1}. So, by considering these values anodic peak current (I_{pa}) *vs.* scan rate brings to close the intention of electron relocate or transfer reaction under a diffusion controlled at the TSC/poly (paraphenylenediamine) electrode surface [31,32].

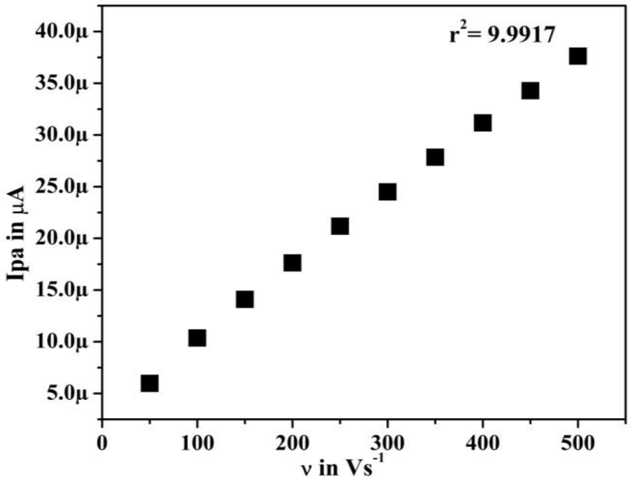


Figure 6. Graph of anodic peak current· of DP versus scan rate

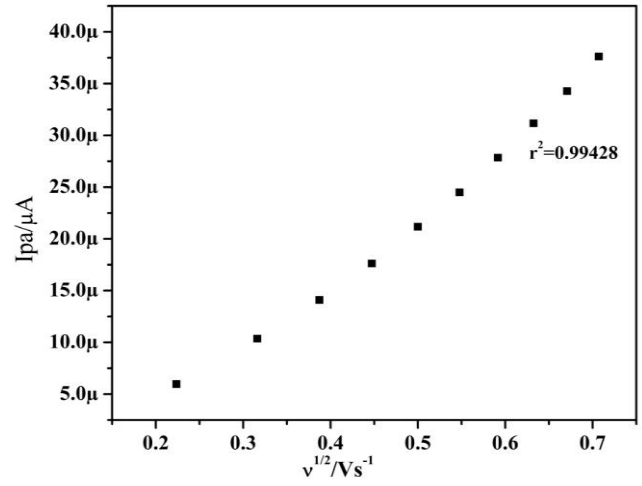


Figure 7. Graph of anodic peak of DP versus square root of scan rate

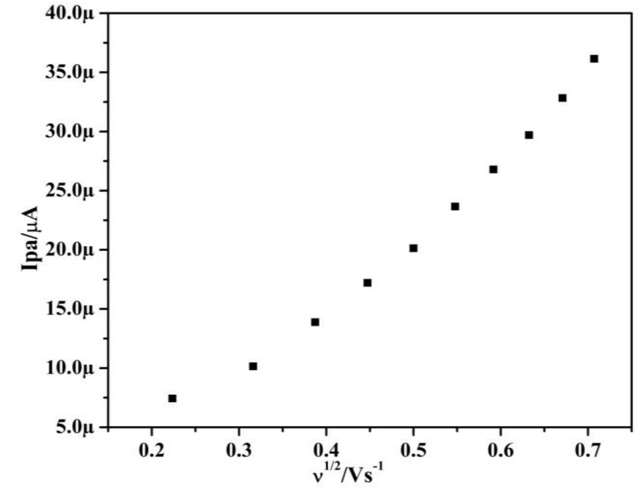


Figure 8. Graph of anodic peak current of UA versus square root of scan rate

3.3.2. Effect of Dopamine and Uric acid Concentration

The effect of concentration of DA and UA of the cyclic voltammograms obtained with the help of different meditations from 1 to 4µM attained by TSC and poly(paraphenylenediamine) MCPE at 0.2 M phosphate buffer· solutions of pH 7.4 at the presence of scan rate 0.1Vs^{-1}, and this is enlightening in the Figure 9, 10, 11 and 12, gives us an idea about that by way of increased concentration, the I_{pa} current values of DP and UA hoists, which· terminates linear relationship concerning of anodic peak current and concentrations. The graph of anodic peak current (I_{pa}) *vs.* concentrations makes a clear linear relationship and equation of linear regression is I_{pa} (µA) = 0.598(C_0 µM) +1.625, (r^2 = 0.9997) and I_{pa} (µA).= 0.6895 (C_0 µM) + 1.539, (r^2 = 0.96152) for DP and UA respectively. The study of limit of detection (LOD). and limit of quantification (LOQ) was premeditated by following equations. Whereas S is the standard deviation of mean value, the working ʿcurve slop is M [31-34,36].

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

$$LOQ = 10\ S/M \quad (3)$$

Pragmatic standards of LOD and LOQ for together DP and UA all along through TSC and poly(paraphenylenediamine) MCPE are list in Table 1, and also Table 1 give explanation the lower limits of detection of DP and UA for TSC and poly(paraphenylenediamine) MCPE than other modified carbon paste electrodes [34-39].

Table 1. Comparison of the detection limits of DP and UA with different modified electrodes with thiosemicarbazide (TSC) and poly(paraphenylenediamine) MGCE

Working Electrode	Limit of Detection (LOD) in µM		Method	References
	Dopamine (DP)	Uric acid (UA)		
Au-RGO/GCE	1.4	1.8	CV	[35]
PdAgNFs/rGO/GCE	0.048	0.081	CV	[36]
Fe_3O_4@AuGA/GCE	0.02	0.05	CV	[37]
AuNPs-β-CD-Gra/GCE	0.15	0.21	CV	[38]
ERGO/GCE	0.5	0.5	CV	[39]
Imprinted PPy/GCE	5.7	------	CV	[40]
TSC/poly(paraphenylenediamine) MGCE	1.3	4.1	CV	This work

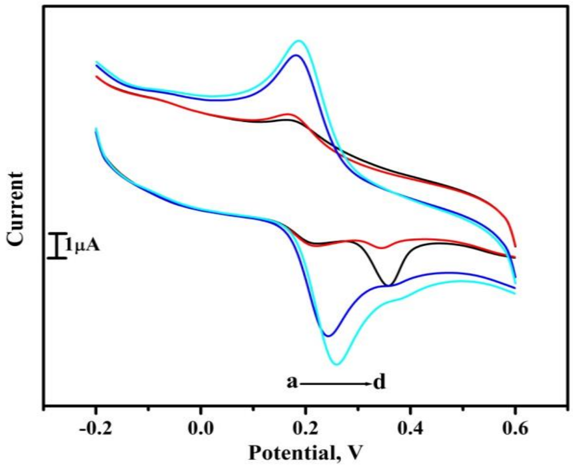


Figure 9. Cyclic voltammograms of DP in 0.2 M PBS solution of pH 7.5 at TSC and poly(paraphenylenediamine) MGCE at scan rate of 0.05 Vs^{-1} with different concentration (a–d; 1 to 4 mM)

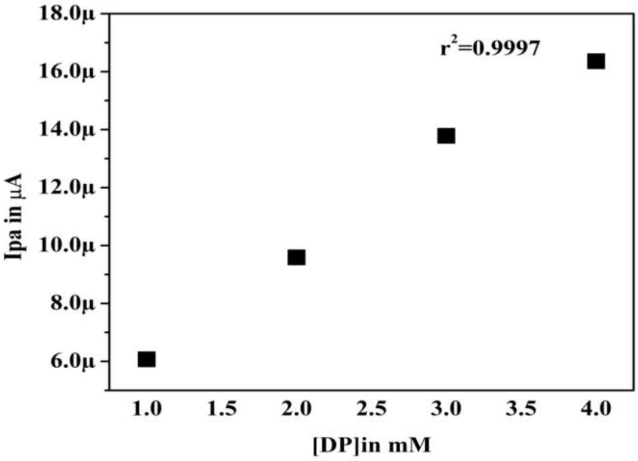


Figure 10. Graph of anodic peak current versus concentration of DP

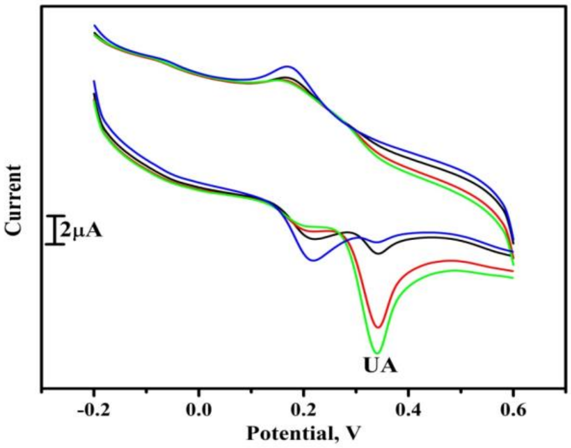


Figure 11. Cyclic voltammograms of UA in 0.2 M PBS solution of pH 7.5 at TSC and poly (paraphenylenediamine) MGCE at scan rate of 0.05 Vs^{-1} with different concentration (a–d; 1 to 4mM)

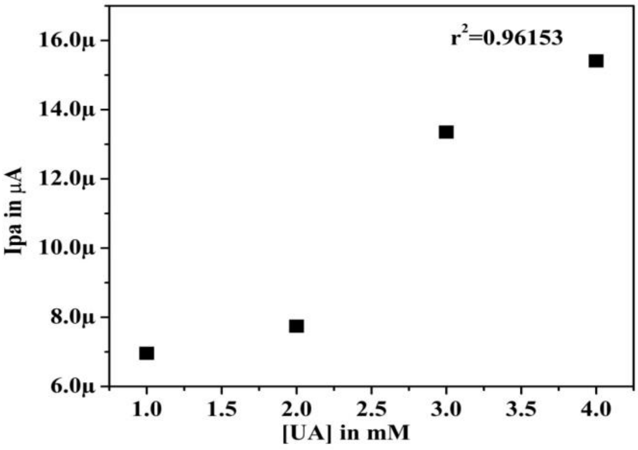


Figure 12. Graph of anodic peak current versus concentration of UA

3.3.3. Influence of pH and Dopamine and Uric acid oxidation

More often than not in electrochemical declaration of composites with the intention of electro active pH of the behind electrolyte plays a momentous role, as shown in Figure 13, 14, 15, and 16, the effect of pH value on the CV retort of the binary mixture including 40 µM DP and 40 µM UA was explore at TSC/poly(paraphenylenediamine) MCPE 0.2 PBS was worn to acquire cyclic voltammograms of DP and UA with varying pH values and also observe anodic peak potentials (I_{pa}) of DP and UA was stimulated to the supplementary negative side as the solution of pH increases. The linear relationship was fashioned stuck between the DP and UA anodic peak potentials *vs.* the pH of PBS in the assortment of 5.5 to 0.8. The equations of linear regression are Epa(V) = 0.08-0.04 (pH) (r^2 =0.9956) and E_{pa}(V) = 0.2-0.3 (pH) (.r^2 = 0.96153) for DP and UA independently. The 1.32 and 1.35 slopes for DP and UA oxidation point towards that comparable integer of protons and electrons are caught up in the redox process which is confirmed by Nernst equation [32-35].

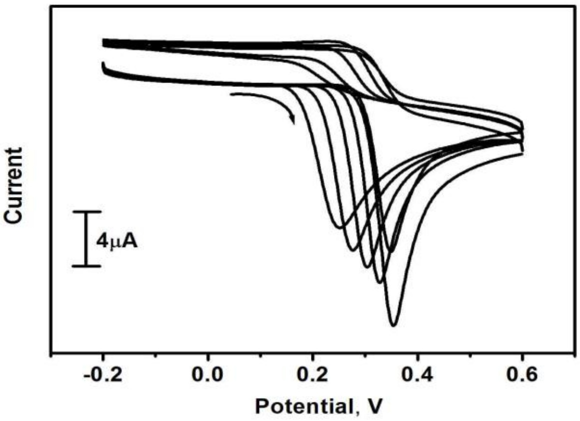


Figure 13. A cyclic voltammograms of 50 µA DP at TSC and poly(paraphenylenediamine) MCPE in 0.2 M PBS solution at different pH

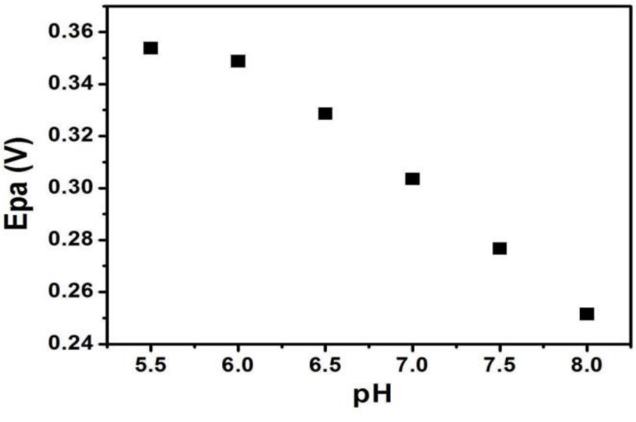


Figure 14. A graph of the anodic peak potential of DP at TSC and poly(paraphenylenediamine) MCPE at pH 5.5 to 8.0

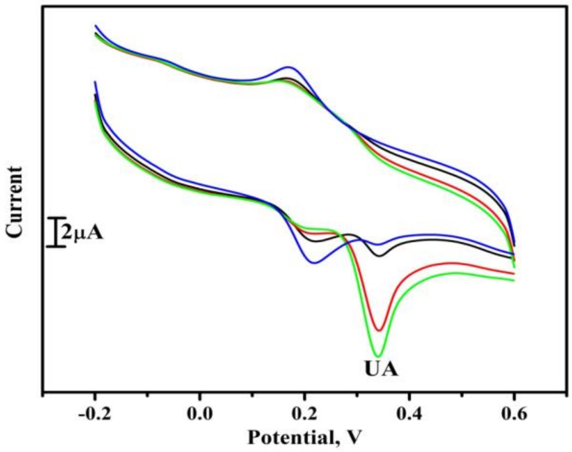


Figure 15. Cyclic voltammograms of UA in 0.2 M PBS solution of pH 7.5 at TSC and poly(paraphenylenediamine) MGCE at scan rate of 0.05 Vs^{-1} with different concentration (a–d; 1 to 4 mM)

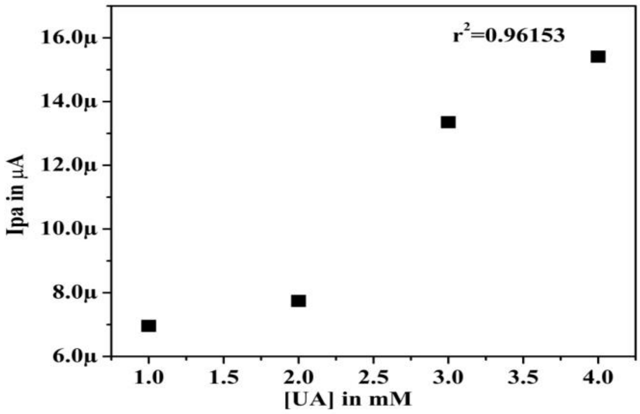


Figure 16. Graph of anodic peak current versus concentration of UA

3.3.4. Intervention study of DP and UA

Thiosemicarbazide (TSC)/poly(paraphenylenediamine) MCPE was second-hand to instantaneous strength of mind to DP and UA, with Differential pulse voltammetry (DPV) for the reason that of its extra current compassion and be deficient in the background current. Figure 17 makes clear DPV confirmation of synchronized analysis of 0.1 mM DP and 0.1 mM UA in 0.2 M PBS of pH 7.4. By this fortitude be approved not in the concentration of solitary analyte was misrepresented and further remnants kept constant. Figure 18 elucidate so as to through augmented concentration, the peak current of DP was elevated, and this awareness improved beginning from 10.0 µM to 40.0 µM, it as well explains to near is refusal distinction in the peak current of UA. The discriminatory oxidation; be pragmatic for D.P at 0.1506 V and for UA 0.3078 V correspondingly, among a peak-to-peak separation’’ was 0.1345V. At last, the results elucidated that accurate and interfering at no cost detection of DP and UA was completed through TSC and poly(paraphenylenediamine) MCPE.

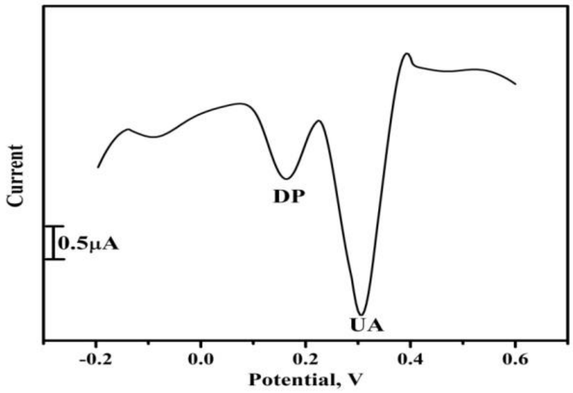


Figure 17. Differential pulse voltammogram obtained for 0.1 mM DP and 0.1 mM UA at TSC and poly(paraphenylenediamine) MCPE

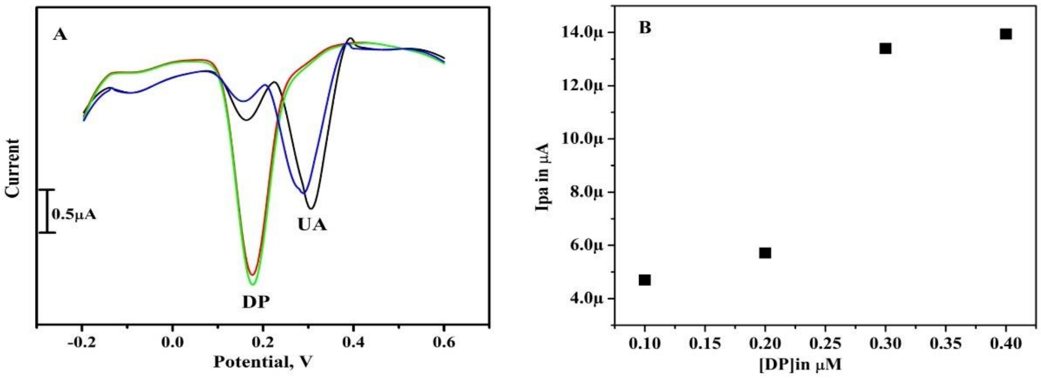


Figure 18. A) Differential pulse voltammograms of (a) 10.0 mM (c) 20.0 mM (d) 30.0 mM (e) 40.0 mM in 0.2 M PBS of pH 7.4 in the presence of 30.0 mM UA at TSC and poly (paraphenylenediamine) MCPE; B) Graph of anodic peak current versus concentration of DP

4. CONCLUSION

The MCPE was prepared by electro ionic deposited with thiosemicarbazide (TSC) and electropolymerized with PMA material and electrochemical parameters are observed by CV method. The modified carbon paste electrode installation was very effective with gaze at to electrochemical investigation of DP and UA. In this TSC/poly(paraphenylenediamine) MCPE expose outstanding impact on redox peak currents for DP and UA with earlier literature. The MCPE constructed incredible impact on redox peak current for DP and UA with enhanced electrochemical warning signs taking apart struck in between DP and UA. The modified electrode has far above the ground electrocatalytic activity with strong selectivity and sensitivity observed in DP and UA concentrations. The high electrocatalytic activity of TSC/

poly(Paraphenylenediamine) MCPE probable to be highly appropriate in the restraint of electro analytical chemistry for willpower of supplementary bioactive molecules or neurotransmitters.

Declarations of interest

The authors declare no conflict of interest in this reported work.

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