Int. J. Electrochem. Sci., 13 **(2018)** 159 – 174, doi: 10.20964/2018.01.40

International Journal of
ELECTROCHEMICAL
SCIENCE
www.electrochemsci.org

Poly(L-Cysteine) Modified Pencil Graphite Electrode for Determination of Sunset Yellow in Food and Beverage Samples by Differential Pulse Voltammetry

Ozge Koyun and Yucel Sahin[*]

Department of Chemistry, Faculty of Arts & Science, Yildiz Technical University, TR34210 Istanbul, Turkey.
[*]E-mail: yucelsahin06@gmail.com

Received: 12 September 2017 / *Accepted:* 29 October 2017 / *Online Published:* 1 December 2017

In this study, we have focused on the use of a poly(L-cysteine) (PLC) modified pencil graphite electrode (PGE) as a sensor for determination of Sunset Yellow (SY). The performance of the modified electrode was studied using cyclic voltammetric and differential pulse voltammetric method. The modified electrode was characterized by electrochemical impedance spectroscopy, cyclic voltammetry and Fourier transform infrared spectroscopy. The surface of the modified electrode was examined by scanning electron microscope. The electrochemical behavior of SY in phosphate buffer solution (pH 7.0) was examined using unmodified PGE and PLC modified PGE (PLC/PGE). The results showed that the electrochemical response of PLC/PGE to SY was significantly developed. The PLC/PGE showed linear responses in the electrochemical oxidation of SY between the concentration values of 1.0 µM and 1000 µM. The sensor (PLC/PGE) showed a good response for SY with a detection limit of 0.125 µM (S/N=3). Analytical application of PLC/PGE was successfully tested in the determination of SY in food and beverage samples.

Keywords: Sunset yellow; Poly(L-cysteine); Electropolymerization; Pencil Graphite Electrode; Food Additive

1. INTRODUCTION

Synthetic food colorant has been commonly used to replace natural food colour in food production industry because of excellent colour uniformity, high stability during preparation processes and low-cost production [1]. Synthetic colorants widely contain aromatic ring structures and azo (N=N) functional group, which may able to adversely affect human health. Sunset Yellow (E110) (SY), is a synthetic azo dye which is widely used in beverages, foods, cosmetics, medicines and

colorings, which can cause migraines, eczema, allergies, diarrhea, anxiety and cancer when over-consumed [2, 3].

The EU and Joint FAO/WHO Expert Committee on Food Additives (JECFA) in 1982 and EU Scientific Committee on Food (SCF) in 1984 has standardized the acceptable daily intake (ADI) for SY between ranges of 0–4 mg/kg bw/day. However, European University Association and several European countries such as Norway and Finland have banned SY to be used in food products [1]. Therefore, the determination of SY in foods is especially important. Until now, numerous analytical methods such as spectrophotometry [4, 5], high performance liquid chromatography [6, 7], thin layer chromatography [8, 9], capillary electrophoresis [10] and enzyme-linked immunosorbent assay (ELISA) [7] have been used for the detection of SY. To overcome the deficiencies of these methods, such as expensive instrumentation, complex sample pretreatment and time-consuming procedures, a quick, appropriate, accurate and sensitive analytical method for SY is immediately sought. As we all know, electrochemical methods are quick, appropriate, highly sensitive and very low cost are environmentally friendly in comparison with other methods.

Until today, different modified working electrodes have developed for the electrochemical determination of SY present in food products [1, 11-14]. As the polymer modified electrode was originally introduced by Miller and some other scientists in 1978, the chemically modified electrode areas were expanded. The chemical and physical stability and three-dimensional structure of polymer films offer effective potential and more reaction sites for the electrode reaction. Research on the application of amino acids polymer modified electrodes has attracted increasing attention in recent years [2, 3]. L-cysteine was used to modify electrodes using chemical and electrochemical methods through its functional groups such as amine, carboxyl, and sulfhydryl. The aim of this work is to develop a sensitive, appropriate and environmentally friendly method for the determination of SY, based on the extraordinary activity of L-cysteine.

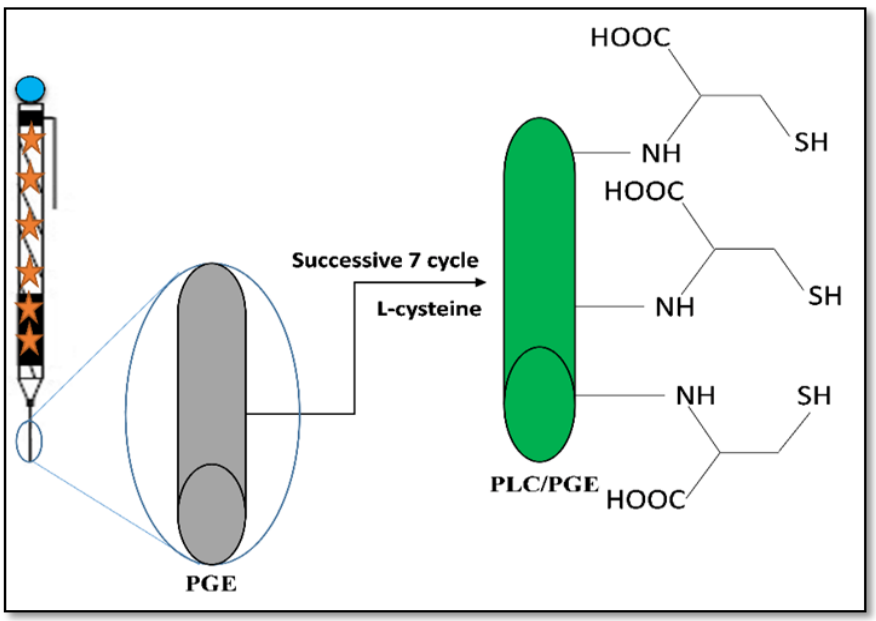


Figure 1. Schematic representation of the preparation of PLC/PGE.

In this study, production, characterization, and analytical performance of the sensor for Sunset Yellow are presented including poly (L-cysteine) on the surface of a PGE. Scheme of the preparation process of PLC/PGE is shown in Fig. 1. Control of the proposed sensing method was made on real food and beverage samples to be applied in the determination of Sunset Yellow for its critical concentration range and toxicity.

2. MATERIALS AND METHODS

2.1. Materials

Sunset Yellow FCF (Dye Content 90%), L-cysteine (97%), Phosphoric acid (85%), Potassium hydroxide (≥85%), Potassium hexacyanoferrate(II) trihydrate (≥99%), Potassium hexacyanoferrate(III) (≥99%) were supplied from Sigma-Aldrich. Potassium dihydrogen phosphate (≥98%) and Dipotassium hydrogen phosphate (≥98%) were obtained from Merck. The solutions of Sunset Yellow were prepared daily. Phosphate buffer solutions (PBS) were prepared from dipotassium hydrogen phosphate and potassium dihydrogen phosphate solutions. Food and beverage samples were supplied from a local patisserie and market. All solutions were prepared using 18 $M\Omega cm^{-1}$ ultrapure water provided with a MILLIPORE Milli-Q Direct 16.

2.2. Apparatus

A five-necked electrochemical glass cell was used in the throughout the experimental procedures. PGE was used as working electrode. An Ag/AgCl (in 3.0 M KCl) (CH Instruments Inc. CHI 111) and a home-made Pt electrode was used as a reference and auxiliary electrodes, respectively. The PGE was formed by cutting the leads (Tombow, HB, D: 0.5mm) into 3cm long pieces and 1.0 cm of each piece was immersed in the solution. Voltammetric measurements were conducted by Autolab PGSTAT 128N (software: Nova 1.11.1) Potentiostat/Galvanostat. Electrochemical impedance spectroscopy (EIS) measurements were carried out by a Gamry Reference 3000 Potentiostat/Galvanostat system. Scanning electron microscopy (SEM) micrographs were obtained with ESEM-FEG Philips XL-30 instrument. Infrared spectra were recorded on a Perkin Elmer Spectrum One FTIR (ATR sampling accessory) spectrophotometer. The absorbance measurements were carried out by an Agile 8453 UV-visible (UV-vis) spectrophotometer.

2.3. Preparation of modified electrode (PLC/PGE)

The electrochemical polymerization was performed in a phosphate buffer solution (0.1 M, pH 4) containing 1.0 mM L-cysteine. In the polymerization process of L-cysteine, the potential scan was cycled 7 times between 2.4 and -0.5 V, at a scan rate of 100 mV/s. After the polymerization process, the electrode was washed with deionized water and was stored in a desiccator until use.

2.4. Preparation of real samples

The cake sample (5.00 g), which was obtained from a local patisserie in Turkey used as a real sample in this work. This sample was prepared as follows: 5.00 g cake was added to 25 mL of PBS pH 7.0 and stirred for 30 min to form homogeneous solutions. Later, cake sample was filtered using a filter paper and stored at +4 °C until analysis.

The jelly sample (1.00 g), which was obtained from a local market in Turkey used as another real sample in this work. This sample was prepared as follows: 1.00 g jelly was added to 25 mL of PBS pH 7.0 and stirred for 15 min to form homogeneous solutions. Later, the jelly sample was filtered using a filter paper and stored at +4 °C until analysis.

Samples of fruit juice commercially available in the local market were without any pretreatment used as real samples in this work. 1.0 mL each of fruit juice samples were diluted with 0.1 M PBS (pH 7.0) solution and ready for analysis.

2.5. Analytical procedure

Voltammetric measurements on PLC/PGE were taken in the electrochemical cell including 25 mL 0.1 M phosphate buffer solution pH 7.0. The differential pulse voltammetry (DPV) under the optimal conditions (initial potential of 0.4 V, final potential of 1.0 V, the scan rate of 100 mV s^{-1}, modulation amplitude of 50 mV, wait time 5 s and step potential of 5 mV) with the consecutive addition of various concentration of SY were used for the calibration curve. Each of the measurement was conducted with a new electrode.

3. RESULTS AND DISCUSSION

3.1. Morphology and characterization studies of PLC/PGE

The characterization and surface morphology of PLC/PGE was performed by cyclic voltammetry (CV), electrochemical impedance spectroscopy (EIS) and Fourier transform Infrared spectroscopy (FT-IR) and scanning electron microscopy (SEM).

The redox species of $Fe(CN)_6^{4-/3-}$ was chosen for the electrochemical characterization study. The CVs of 2.5 mM $Fe(CN)_6^{4-/3-}$ that were obtained by PGE and PLC/PGE and are indicated in Fig. 2. We have detected two peaks by PGE at 0.27 V and 0.14 V. Potential difference (ΔE) between the anodic and cathodic peaks value of 0.13 V. The ΔE value of these peaks at 0.25 V and 0.13 V was 0.12 V with PLC/PGE (Fig. 2). Accordingly, we can say that the electron transfer rate in redox processes increases in the presence of PLC on PGE. Moreover, we have determined that the increase in the peak currents of $Fe(CN)_6^{4-/3-}$ is an indicator of increasing surface area of the electrode in the case of PLC/PGE. We have discovered that the positively charged functional groups on pencil graphite electrode surface that were formed as a result of the electropolymerization attracted $Fe(CN)_6^{4-/3-}$.

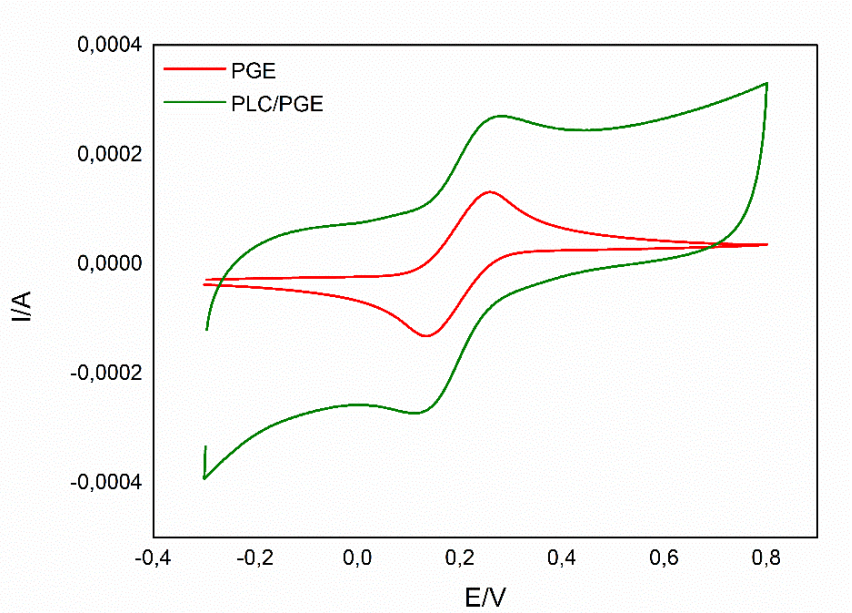


Figure 2. Cyclic voltammograms of PGE and PLC/PGE in 0.1 M KCl solution containing 2.5 mM $Fe(CN)_6^{4-/3-}$ scan rate 50 mV s^{-1}.

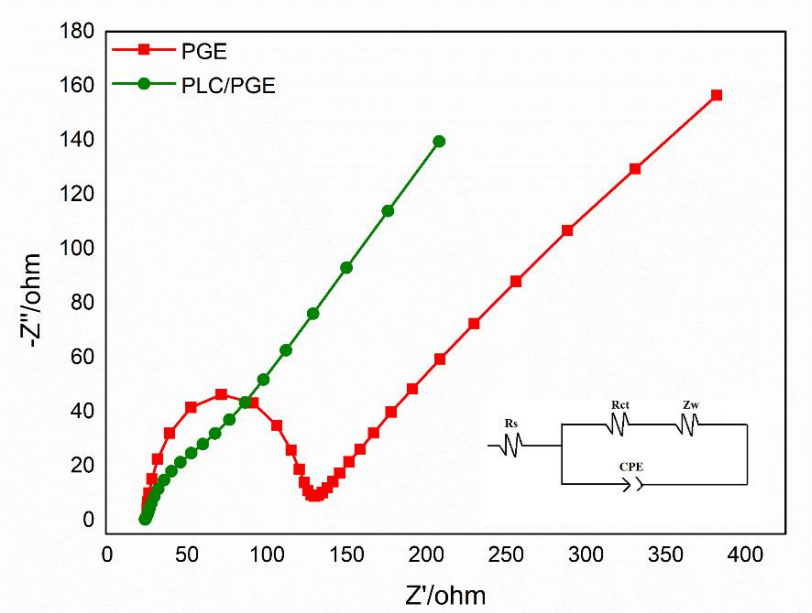


Figure 3. Nyquist diagrams of PLC/PGE and bare-PGE in electrolytes consisting of 1.0 mM $K_3[Fe(CN)_6]$ + 1.0 M KCl; inset: used equivalent circuit model for bare-PGE fitting and PLC/PGE fitting of spectra.

EIS can supply beneficial information on the impedance changes of the electrode surface to characterize the construction process of the sensor. The capacity of electron transfer of bare PGE and PLC/PGE electrodes was also examined using EIS, as shown in Fig. 3. Nyquist diagrams of PLC/PGE and bare-PGE were carried out in 1.0 mM $K_3[Fe(CN)_6]$ + 0.1 M KCl solution at room temperature. The spectra were fitted to a known equivalent circuit model, shown in Fig. 3 (inset), which was widely reported in the literature [2, 15]. The higher electrocatalytic behavior of PLC/PGE was proven by the reduction in charge transfer resistance (Rct). By fitting the data using a suitable equivalent circuit, the

values of charge transfer resistance were determined to be 105 Ω, 63.61 Ω bare-PGE and PLC/PGE, respectively. These results demonstrated higher electron transfer rate and capacities of PLC/PGE.

FTIR spectroscopy was used in the characterization of functional groups of PLC formed on the electrode surface. Characteristic IR bands of PLC/PGE are shown in Fig. 4. The bands belonging to the functional groups are given in sequence (ATR, 4000-600 cm^{-1}): The vibration band for the S-H group is at 2543 cm^{-1}. This characteristic band shows that L-cysteine accumulates on the PGE electrode surface by polymerization. In addition, the bands at 1576 cm^{-1} and 1414 cm^{-1} belong to the $a_{symmetric}$ (COO) and $s_{ymmetric}$ (COO) functional groups, respectively. The band at 1110 cm^{-1} belongs to the S-H functional group [16].

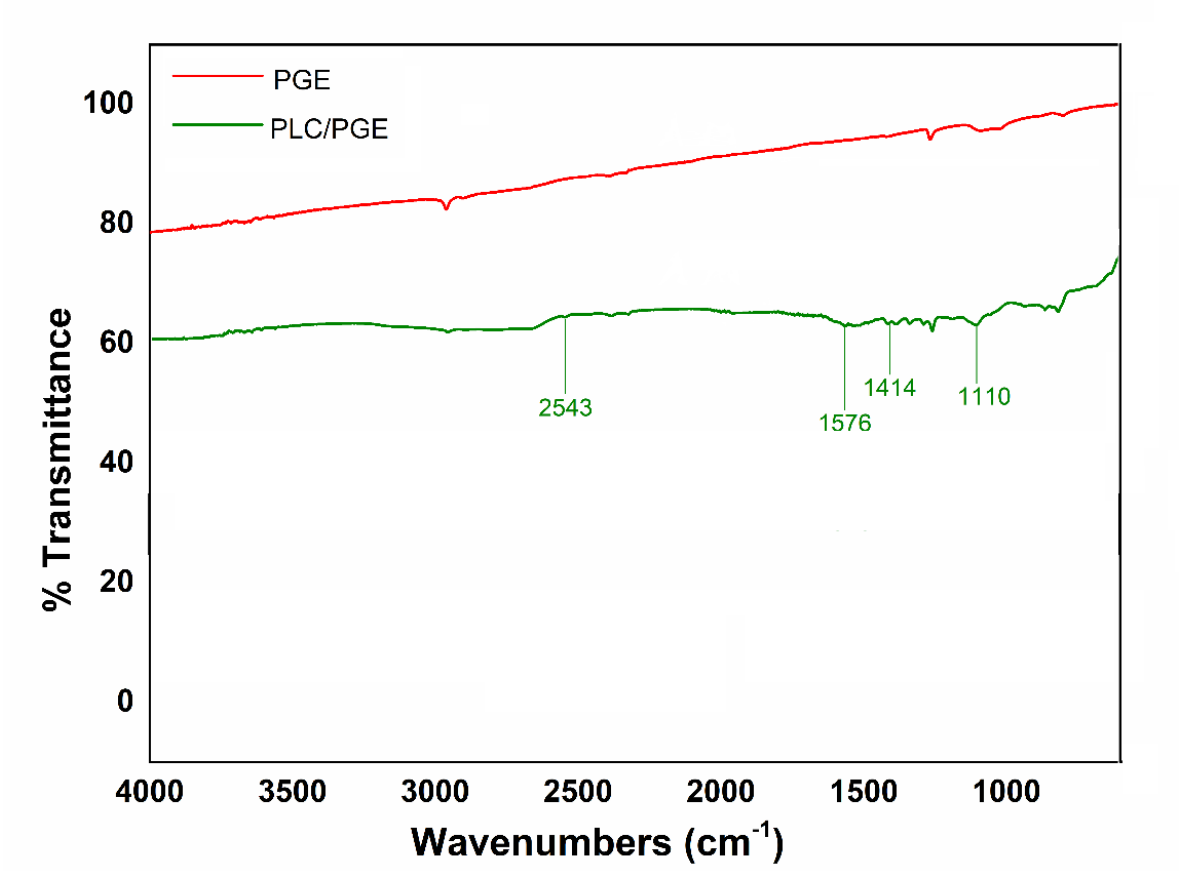


Figure 4. FT-IR spectra of the Bare-PGE and the PLC/PGE.

The surface morphology of the electrodes was investigated by SEM. The bare-PGE appears flat structure as shown in Fig. 5a. When PGE is modified with PLC, sphere-like structure morphology was observed for PLC. The electrode surface becomes rough due to the adherence of the PLC film to the PGE. Fig. 5b and 5c show the PLC on the PGE. This clearly confirms that the L-cysteine has polymerized onto the PGE surface. The sphere-like structure morphology of PLC with its rough surface increases the contact area which in turn can display improved electrochemical behavior. Therefore, it can be assumed that the prepared electrode was PLC/PGE.

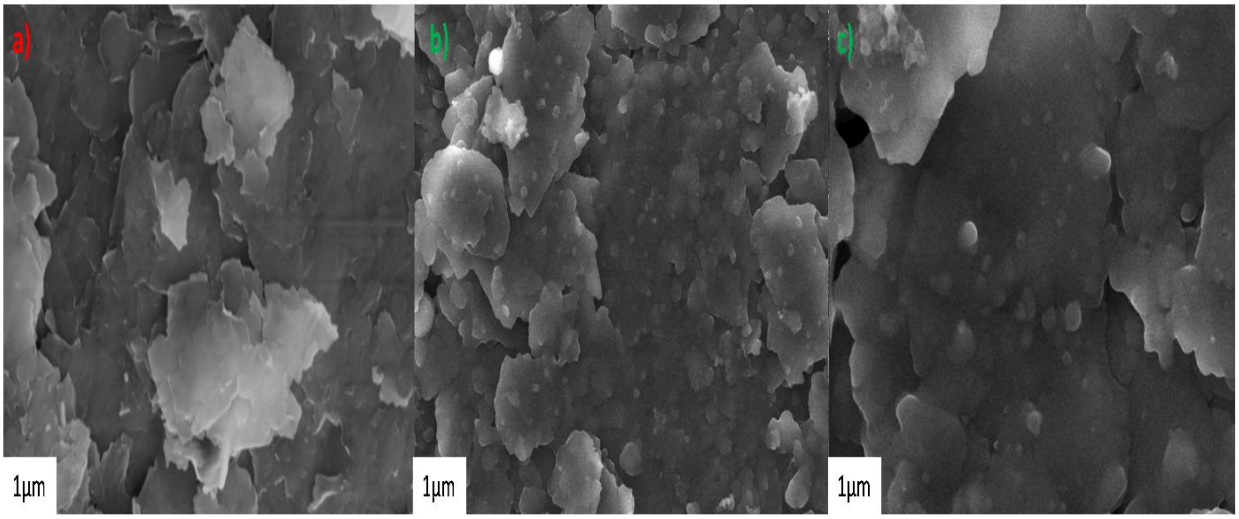


Figure 5. Scanning electron microscopy micrographs of the **a)** Bare-PGE (with 25000 magnification), **b)** PLC/PGE (with 25000 magnification) and **c)** PLC/PGE (with 50000 magnification).

3.2. Calculation of electroactive surface area

We have measured the peak current at different scan rates by which the electroactive surface areas of the electrodes (PGE and PLC/PGE) were defined using the Randles-Sevcik equation, via CV in a 0.1 mol L^{-1} KCl solution + 1.0 mmol L^{-1} $K_4Fe(CN)_6$ [15]. We used the Randles-Sevcik equation to determine the dependence of peak current Ip on the scan rate $\nu^{1/2}$ (1):

$$Ip = (2.69x10^{-5})\, n^{1/2}\, AD^{1/2}\, C^{*}\nu^{1/2} \quad (1)$$

In the equation, n denotes the number of electrons that involve in the redox reaction, v represents the scan rate of the potential perturbation (V/s), A shows the area of the electrode (cm^2), D denotes the diffusion coefficient of the molecules in the solution (cm^2 /s), C* indicates the concentration of the probe molecule in the bulk solution (mol cm^{-3}), and Ip defines the peak current of the redox couple. According to the equation, we can calculate the effective surface area (A) using the value of Ip / $v^{1/2}$, while maintaining the constant values of D, n (n = 1), and C* (1.0 mol cm^{-3}). The diffusion constant value at 25 °C (D= 6.7 × 10^{-5} cm^2 s^{-1}) was determined using the previously published data [15,17]. On both electrodes, a linear increase in the Ip was observed as the square root of the potential scan rate ($v^{½}$) increased, which indicates the reversibility of the reactions occurring on the modified electrode. The electroactive areas of the PGE and PLC/PGE were calculated as 0.0018 cm^2 and 0.0045 cm^2, respectively. Therefore, we see that as the bare PGE surface was modified, the electroactive surface area increased, thus, the sensor (PLC/PGE) became more sensitivity to Sunset Yellow.

3.3. Electropolymerization of L-cysteine

The PLC was accumulated on the bare pencil graphite electrodes by electropolymerization process in 0.1 M phosphate buffer solution (pH 4) containing L-cysteine (1.0 mM) using cyclic

voltammetry (CV). In polymerization process, the electrode was coated with an L-cysteine film and one of the two redox couples was negative. This peak can be classified as a monomer type redox activity. In addition, the other broad peaks indicate the characteristics of polymer semiconductor film peaks. In the electrodeposition of polymer, as the first signal given to a monomer at -0.74 V decreased and the successive cycles increased the peaks corresponding to the polymer. This is due to the growth of an electrochemically active coating (Fig.6). 7 cycles were applied for electropolymerization of L-cysteine.

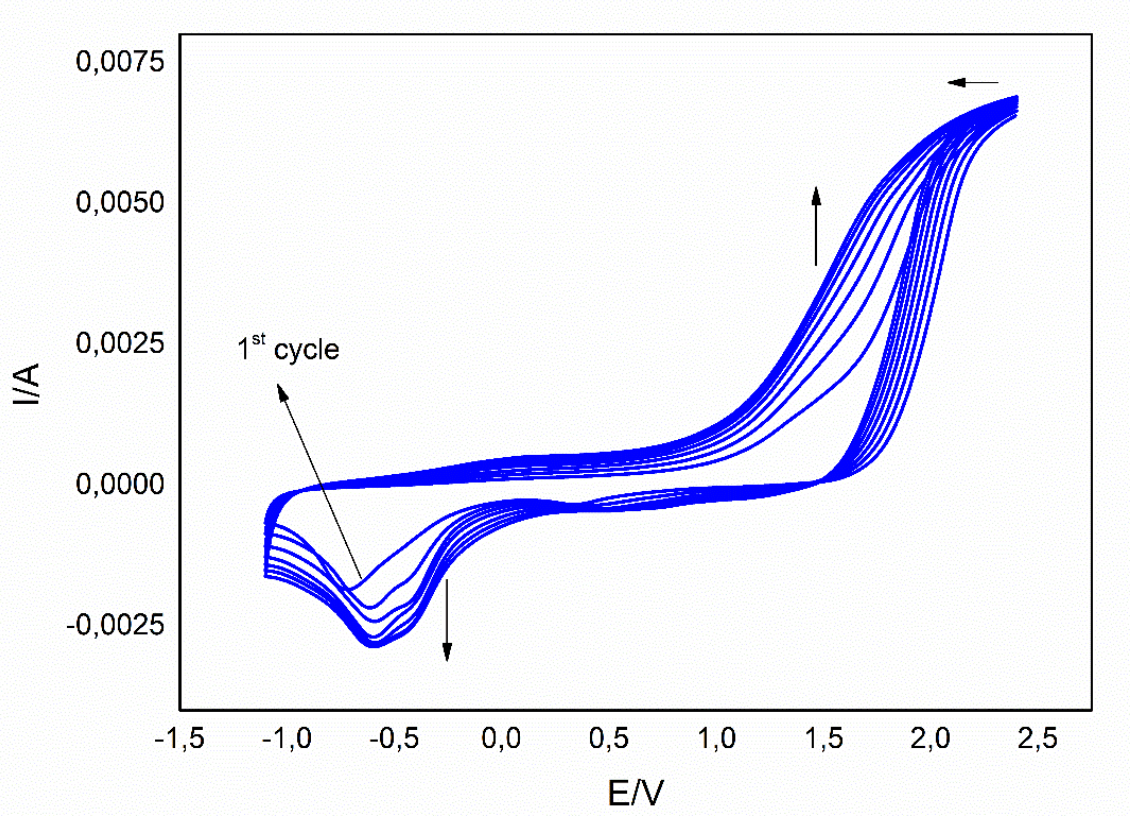


Figure 6. The growth of poly(L-cysteine) film on pencil graphite electrode by potential cycling at a scan rate of 100 mV/s. Solution contains 1.0 mM L-cysteine in 0.1 M PBS (pH 4.0).

3.4. Effect of the number of cycles

Determining of Sunset Yellow was conducted by DPV due to the low sensitivity of CV. Therefore, the effect of number of cycle on the peak current of SY was investigated. The number of cycle examination showed that the peak current considerably increased by increasing the number of cycle from 1st cycle to 7th cycle. After 7 cycles the peak current of SY was decreased (Fig.7). Higher cycles lead to more extensive electropolymerization, and therefore to the formation of thicker sensing film with less accessible sites. Therefore, the optimum number of cycle was chosen as 7 for further studies.

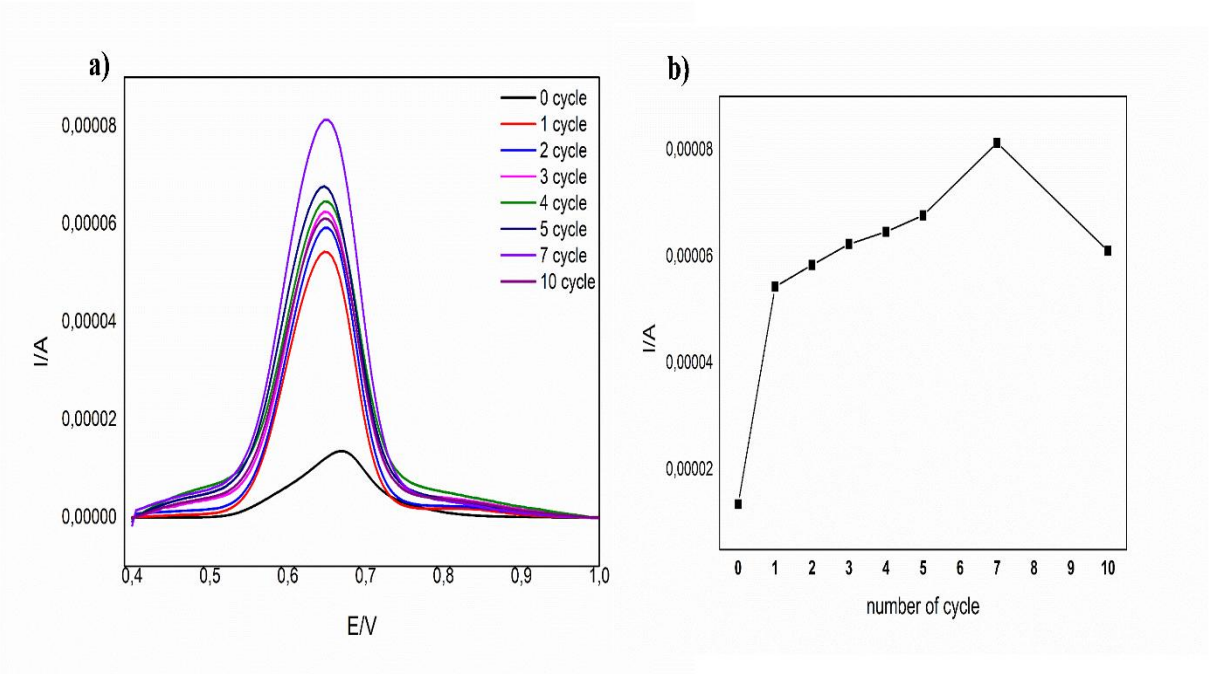


Figure 7. a) Superimposed DPV of SY obtained at different number of cycle of PLC/PGE in 0.1 M PBS (pH 7.0). **b)** Effect of the number of cycle on the peak current of 1.0 mM Sunset Yellow.

3.5. Electrochemical studies

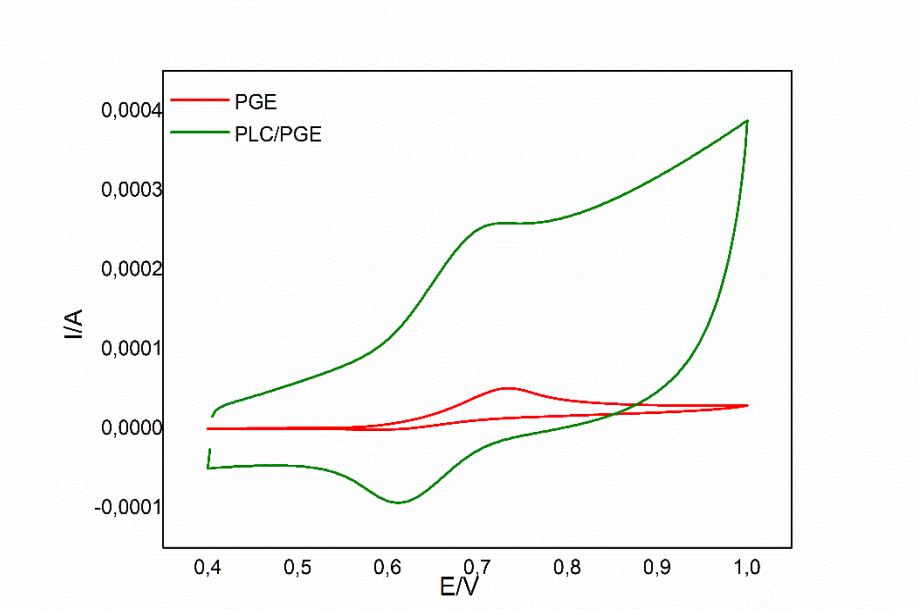


Figure 8. Cyclic voltammograms of phosphate buffer solution (0.1 M, pH 7.0) containing 1.0 mM Sunset Yellow at the bare PGE and PLC/PGE, using the scan rate of 100 mV/s.

Electrochemical oxidation and reduction of SY on the bare PGE and modified PGE were examined by CV technique in PBS (pH 7.0) containing 1.0 mM SY at the scan rate of 100 mV/s. The recorded cyclic voltammograms (Fig. 8) of electrodes were compared for the sensitivity of the modifier film. In the potential range of 0.4 to 1.0 V, one oxidation and one reduction peaks were

observed at the surface of PLC/PGE. Besides, a defined peak at 730 mV appeared at the bare PGE that is because of oxidation of SY. Electropolymerization of L-cysteine on the PGE was associated with peak current increasing and sensitivity that increases the conductivity and surface area of PLC/PGE.

3.6. Effect of scan rate and pH on electrochemical behavior of Sunset Yellow

Cyclic voltammetry was performed at different scan rates for the 1.0 mM Sunset Yellow solution. The measurements carried out in the range of 5–250 mVs^{-1} showed that there is a linear relationship between the square root of the scan rate ($v^{1/2}$) and the peak current ($\dot{I}p$) in the scan rates of 5-250 mVs^{-1} for SY. This indicates that the existence of a diffusion-controlled mechanism prevails for SY (Fig. 9) [15]. The linear regression equations of SY are expressed as follows (2):

$$\dot{I}p = 3x10^{-5}\, v^{1/2} - 4x10^{-5}, \quad R^2 = 0.9906 \qquad (2)$$

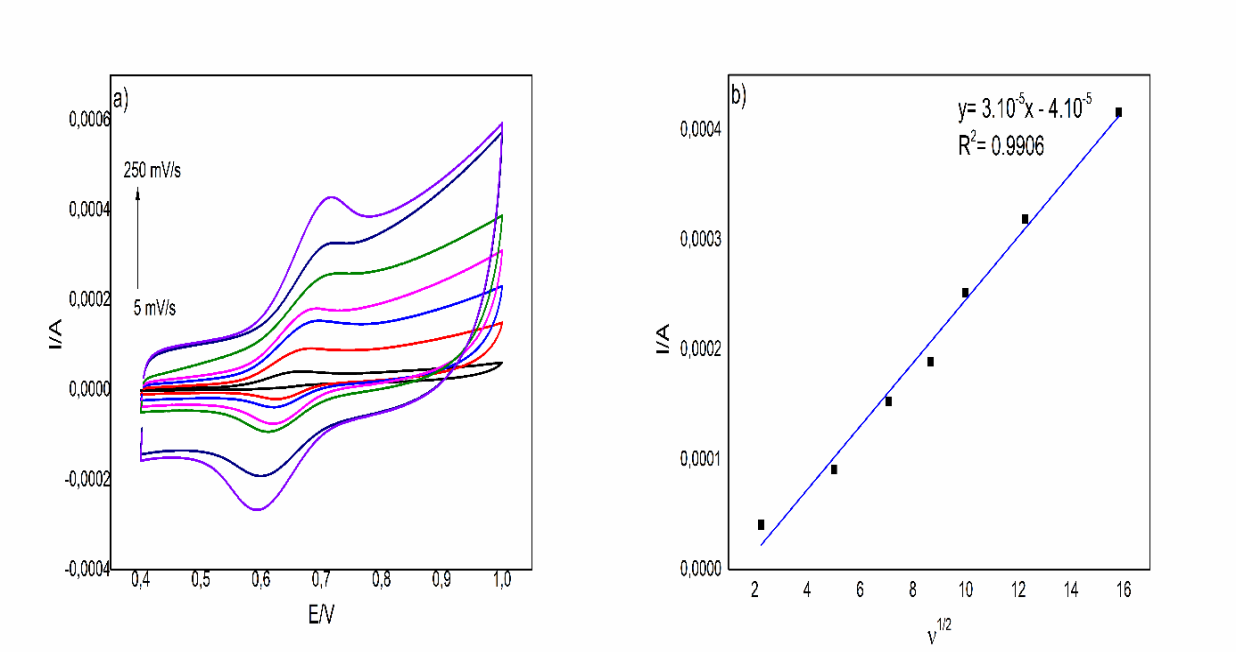
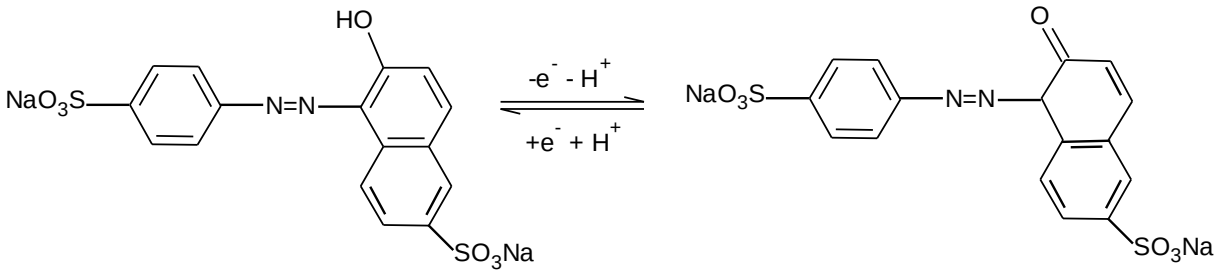


Figure 9. a) CVs of the PLC/PGE in 0.10 M PBS (pH 7.0) containing 1.0 mM Sunset Yellow at different scan rates. b) The linear dependence of I_p on $v^{1/2}$.

The effect of different pH values on SY oxidation at PLC/PGE was examined using DPV in 0.10 mol L^{-1} PBS within pH range of 4.0-8.0. As illustrated in Fig. 10a, as the pH value approached to 7.0, the peak current corresponding to the SY oxidation also increased, and however, when the pH value above 7.0, the peak current started to decrease (see Fig. 10b). In the conditions of neutral media, SY may have the following transformation:



Sunset Yellow is a protic aromatic molecule, which easily undergoes deprotonation and turns to anions at high pH, resulting in the electrostatic repulsion between SY and PLC/PGE [1]. Therefore, for this study, we defined the optimum electrolyte as the PBS of pH 7.0 for the electrochemical detection of Sunset Yellow.

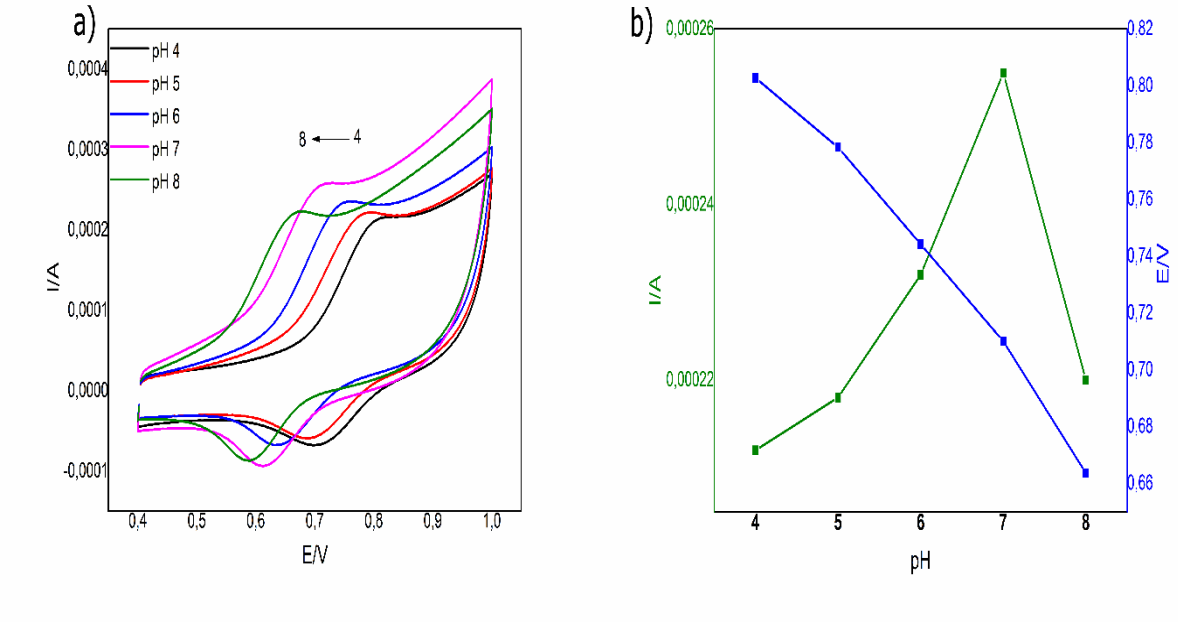


Figure 10. a) DPVs of the PLC/PGE in 0.10 mol L^{-1} PBS containing 1.0 mM Sunset Yellow at different pH values. **b)** The relationship between current-potential-pH values.

3.7. Analytical application

The limit of detection and limit of quantitation the effect of Sunset Yellow concentration under optimized parameters on the behavior of the PLC/PGE was examined using DPV method in order to obtain the dynamic range (Fig. 11). The modified electrode shows working concentration range of 1.0-1000 µM with linear regression equation (3):

$$Ip\,(A) = 0.0814\,C_{SY}(M) - 9x10^{-8}, \qquad R^2 = 0.9998 \qquad (3)$$

The limit of detection (LOD) and limit of quantitation (LOQ) were calculated using the relation kS/m, where k=3 for LOD and 10 for LOQ, S representing the standard deviation of the peaks current

of the blank (N = 10) and m representing the slope of the calibration curve. LOD and LOQ values were found to be 0.125 μM and 0.417 μM, respectively.

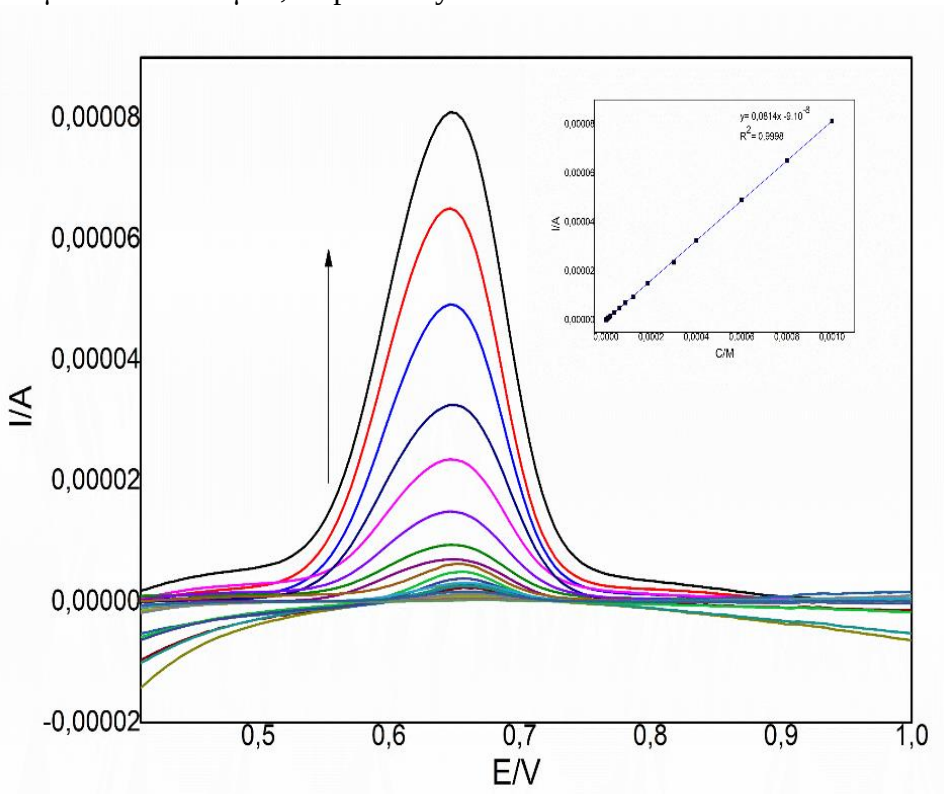


Figure 11. DPVs and calibration curve (inset) for Sunset Yellow at optimal conditions: Applied potential: (0.4 V) - (1.0 V), modulation amplitude of 50 mV, wait time 5 s, step potential of 5 mV and scan rate of 100 mV/s.

These results show the sensitivity of the prepared sensor and proposed method. Table 1 provides a comparison of some studies related to various electrodes used for the determination of SY. The proposed modified electrode have unique properties in comparison with reported other modified electrodes [2, 3, 11, 18-23]. Firstly, the PGE based sensor for SY was used the first time by this study in the literature. In this case, the prepared sensor offers some advantages such as easy preparation, low-cost, disposable and environmentally friendly. In addition, it has a quite wide linear range. In this way, the prepared sensor allows to work with a large number of real sample types. Finally, satisfactory recovery results were obtained in the determination of SY in cake, jelly and fruit juice samples, indicating that the PLC/PGE can be successfully used to determine the additives in these types of food and beverage samples. Although works by [11], [20] and [23] show very low LOD values, linear ranges are very narrow. The performance of the method proposed by [19] is comparable to our method; however, electrode used in this method was rather complicated and preparation process is time consuming. Further, HMDE used by [21] is hazardous due to the toxicity of volatile mercury. The performance of the method developed by [2], [3] and [22] is comparable to our method; however, electrodes used in these methods were more costly.

Table 1. Comparison of the analytical parameters of the proposed electrode with other reported sensors.

Electrode	Method	Linear range (μmol L^{-1})	Detection limit (μmol L^{-1})	Ref.
poly-L-cys/GCE	DPV	0.008-0.7	0.004	[2]
PLC/Ag/GCE	DPV	0.5-300	0.075	[3]
Au/CPE	DPV	0.1-2	0.03	[11]
MWCNT/GCE	DPV	0.37-75	0.188	[18]
ZnO/Cysteic acid/GCE	DPV	0.1-3.0	0.03	[19]
GO-MWCNT/GCE	DPV	0.09-8	0.025	[20]
HMDE	ASV	0.007-0.35	0.0035	[21]
PLPA/GCE	DPV	0.4-14	0.04	[22]
PDDA-Gr-Pd/GCE	DPV	0.01-10	0.002	[23]
PLC/PGE	DPV	1-1000	0.125	This work

Abbreviations: DPV: differential pulse voltammetry; ASV: adsorptive stripping voltammetry; GCE: glassy carbon electrode; CPE: carbon paste electrode; HMDE: Hanging mercury drop electrode; MWCNT: multi-walled carbon nanotube; ZnO: zinc oxide; GO: graphene oxide; PLPA: poly(L-phenylalanine); PDDA: poly(diallyldimethylammonium chloride); Gr-Pd: graphene-palladium nanoparticle; PLC: poly(L-cysteine).

3.8. Reproducibility and stability of PLC/PGE

The reproducibility of the prepared modified electrode for 0.1 mM SY was examined. Peak current responses were determined via 10 electrodes, which were produced under the same conditions. Response peak intensity shows that the results are reproducible with a relative standard deviation of 2.97%. The stability of the electrode was controlled by its storing in desiccator in the room temperature for 90 days and then the response of electrode was examined for the determination of 0.1 mM SY solutions. The response of the PLC/PGE was about 96% of its initial response. These results show the reproducibility and good stability for the proposed sensor in the determination of SY.

3.9. Interference studies

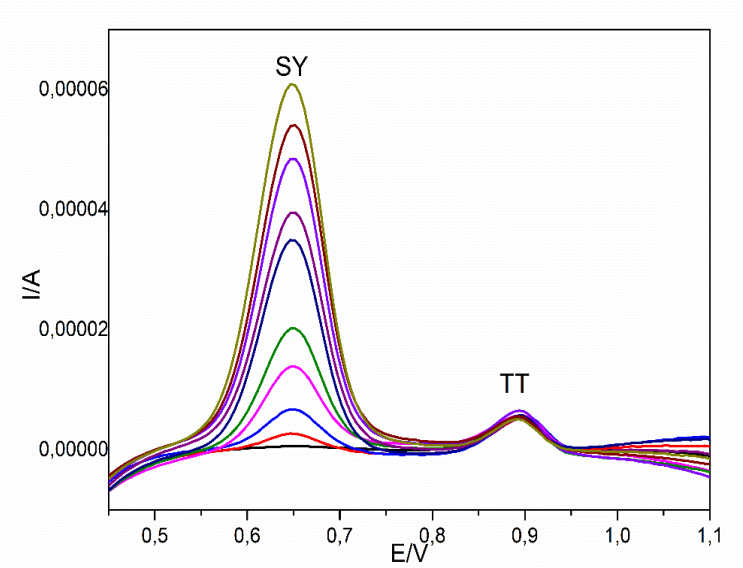


Figure 12. Superimposed DPV of different SY concentrations obtained at PLC/PGE in 0.1 M PBS (pH 7.0) in the presence of constant TT concentrations (0.3 μM).

Firstly, for the Tartrazine, which is frequently studied in the literature, a different amount of Sunset Yellow was added to the 0.3 μM Tartrazine solution in the pH 7 buffer solution, indicating that Tartrazine did not have a serious interference effect on the Sunset Yellow (Fig. 12). Possible interference for the determination of Sunset Yellow on PLC/PGE was examined by addition of different species into 0.1 M PBS solution (pH 7.0) containing 1.0 mM SY. As Fig. 13 showed, most of the species, such as Glucose (G), Tartrazine (TT), Ascorbic Acid (AA) and Quinoline Yellow (QY) in a 100-fold concentration, Ca^{2+}, Cu^{2+}, Zn^{2+}, SO_4^{2-} and NO_3^- in a 1000-fold concentration had a little interference toward the detection of SY. All results suggest that the PLC/PGE has a good anti-interference capability for the detection of SY.

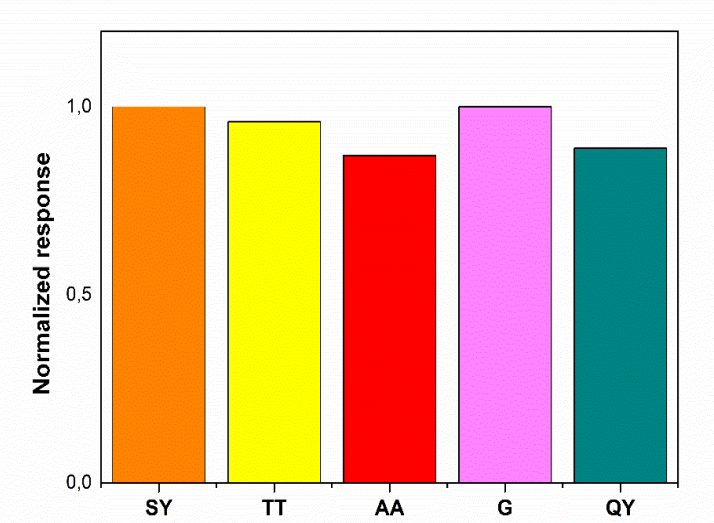


Figure 13. Interference study on the response of 100-fold TT, AA, G, and QY.

3.10. Determination of Sunset Yellow in real samples

In order to confirm the application potential and applicability of the proposed method for the analysis of Sunset Yellow which was used as an additive and coloring in food and beverage products, the PLC modified PGE was applied to the determination of Sunset Yellow in the cake, jelly and fruit juice samples. The real samples were prepared as described in the section of preparation of the real samples. Subsequently, the filtrate was mixed with 0.1 M phosphate buffer solution (pH 7.0) for the determination of SY. Table 2 shows the results obtained by using the PLC/PGE as well as by UV-vis spectrophotometry for the detection of Sunset Yellow in the cake, jelly and fruit juice samples with different concentrations. The recovery using the proposed sensor was from 94.7% to 105.6% in the cake sample, was from 96.4% to 102.3% in the jelly sample and was from 96.4% to 102.3% in the fruit juice samples, which indicated that the prepared PLC/PGE could be satisfactorily used for the determination of SY in the cake-like and jelly and fruit juice samples.

Table 2. Determination of Sunset Yellow in food and beverage samples with PLC/PGE.

Sample	UV-vis Method[a]	Proposed Method[b]	Added	Total after addition[b]	RSD[b] (%) (n=5)
	($x10^{-3}$/mol L^{-1})	($x10^{-3}$/mol L^{-1})	($x10^{-3}$/mol L^{-1})	($x10^{-3}$/mol L^{-1})	
Cake	0.102	0.107	0.1	0.211	3.61
Jelly (orange)	0.219	0.224	0.1	0.329	3.12
Orange juice	0.116	0.119	0.1	0.221	2.94
Mixed fruit juice	0.214	0.219	0.1	0.324	3.11

Abbreviations: [a] The method with UV-vis spectrophotometry was used. [b] The proposed method with PLC/PGE was used.

4. CONCLUSIONS

This work demonstrates that a sensitive sensor for Sunset Yellow was improved using PLC coated PGE. The fabrication of the modified pencil graphite electrode was conducted in the easy electropolymerization method. The Sunset Yellow was determined in a low-cost, fast and reliable way in this study. The application of PLC/PGE was successfully examined in the determination of SY in food and beverage samples. It can be concluded that the prepared PLC/PGE is preferable electrochemical sensor for the voltammetric determination of SY because of its properties; very low cost, single-use character, easy preparation, good sensitivity and selectivity.

ACKNOWLEDGEMENT
We express our sincere thanks to financial support of the Yildiz Technical University Scientific Research Projects Coordination Department (Project No: FDK-2017-3130).

References

1. K. Rovina, S. Siddiquee and S.M. Shaarani, *Food Control*, 82 (2017) 66.
2. K. Zhang, P. Luo, J. Wu, W. Wang and B. Ye, *Anal Methods*, 5 (2013) 5044.
3. Y. Tang, Y. Wang, G. Liu and D. Sun, *Indian J. Chem.*, 55 (2016) 298.
4. L. Capitán-Vallvey, M.D. Fernández, I. de Orbe and R. Avidad, *Analyst*, 120 (1995) 2421.
5. L.F. Capitán-Vallvey, N.N. Iglesias, I. de Orbe Payá and R.A. Castaneda, *Microchim. Acta*, 126 (1997) 153.
6. F.E. Lancasterand and J.F. Lawrence, *Food Addit. Contam.*, 16 (1999) 381.
7. C. Tatebe, T. Ohtsuki, N. Otsuki, H. Kubota, K. Sato, H. Akiyama and Y. Kawamura, *Am. J. Anal. Chem.*, 3 (2012) 570.
8. R. Hoodless, K. Pitman, T. Stewart, J. Thomson and J. Arnold, *J. Chromatogr. A*, 54 (1971) 393.
9. F. Soponar, A.C. Moţ and C. Sârbu, *J. Chromatogr. A*, 1188 (2008) 295.
10. H.Y. Huang, C.L. Chuang, C.W. Chiu and M.C. Chung, *Electrophoresis*, 26 (2005) 867.
11. S.M. Ghoreishi, M. Behpour and M. Golestaneh, *Food Chem.*, 132 (2012) 637.
12. R.A. Medeiros, B.C. Lourencao, R.C. Rocha-Filho and O. Fatibello-Filho, *Talanta*, 97 (2012) 291.
13. C. Qin, W. Guo, Y. Liu, Z. Liu, J. Qiu and J. Peng, *Food Anal. Methods*, 10 (2017) 2293.
14. P. Sierra-Rosales, C. Toledo-Neira and J. Squella, *Sens. Actuators* B *Chem.*, 240 (2017) 1257.
15. O. Koyun, H. Gursu, S. Gorduk and Y. Sahin, *Int. J. Electrochem. Sci.*, 12 (2017) 6428.
16. S.R. Ankireddyand and J. Kim, *Int. J. Nanomedicine*, 10 (2015) 113.
17. S. Konopka and B. McDuffie, *Anal. Chem.*, 42 (1970) 1741.
18. W. Zhang, T. Liu, X. Zheng, W. Huang and C. Wan, *Colloids Surf. B Biointerfaces*, 74 (2009) 28.
19. P.S. Dorrajiand and F. Jalali, *Food Chem.*, 227 (2017) 73.
20. X. Qiu, L. Lu, J. Leng, Y. Yu, W. Wang, M. Jiang and L. Bai, *Food Chem.*, 190 (2016) 889.
21. M. Gómez, V. Arancibia, C. Rojas and E. Nagles, *Int. J. Electrochem. Sci.*, 7 (2012) 7493.
22. M. Chao and X. Ma, *Food Anal. Methods*, 8 (2015) 130.
23. L. Yu, H. Zheng, M. Shi, S. Jing and L. Qu, *Food Anal. Methods*, 10 (2017) 200.