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An Electroanalytical Determination of Sunset Yellow in Food Product by Amplified Nanostructure Carbon Paste Electrode as Sensor

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A new and sensitive food analytical sensor was fabricated using the amplification of paste electrode (CPE) by CdO-SWCNTs and 1-methyl-3-butylimidazolium bromide (MBIZLB), which was then used to determine Sunset yellow (SY) in aqueous solution. The CPE/MBIZLB/CdO-SWCNTs showed excellent catalytic activity for oxidation of SY and improved its current activity up to about 6.12 times. FESEM image of CdO-SWCNTs nanocomposite displayed decoration of CdO nanoparticles at the surface of carbon nanotubes with a diameter about 30 nm. On the other hand, square wave voltammograms of SY showed a linear dynamic range 0.0006-180 µM with the detection limit of 0.3 nM using CPE/MBIZLB/CdO-SWCNTs as the working electrode. Finally, the CPE/MBIZLB/CdO-SWCNTs was used for the determination of SY in juices samples by applying standard addition method with recovery range of 98.88 to 101.84%.

Keyword: CdO-SWCNTs, Composite, sunset yellow, modified sensor

1. INTRODUCTION

Measuring food additives, especially azo dyes, is one of the most important issues in examining the health and quality of food products [1]. Due to the known side effects of the excessive consumption of azo dyes in food samples, its determination in food products has become an important issue in food industry. Therefore, various analytical methods have long been used for the determination of food additives [2-5].

Sunset Yellow is one of the famous azo dyes, which has a wide range application as a coloring additive for beverages and various foods such as soups, sauces, confectionary, cheeses, desserts, snacks, savory, and preserved fruits [6]. Due to its effect on children's behavior, more attention has been paid to the sunset yellow in food industry. Due to the above-mentioned points and the widespread use of Sunset Yellow in food industry, the concentration of this color compound in food samples has become particularly important in order to evaluate the quality of food products [7].

Among the methods used for Sunset Yellow analysis, electrochemical methods have received more attention compared to other techniques [8]. This may be due to the more advantages of electrochemical sensors such as high analysis speed and the ability to modify electrochemical sensors properly for the selective measurement of pharmaceutical and food compounds [9-20]. On the other hand, a wide range application of mediators in modification process and their synergic effects on modification process, create a high sensitivity condition for the fabrication of new types of electrochemical sensors [21-30].

Nanomaterials created a new approach to materials science and surprised science with their unique properties [31-45]. Due to the extraordinary properties of nanomaterials, they were rapidly applied in various branches of science [46-61]. The high electrical conductivity of nanomaterials is one of their unique features, which makes them as a suitable option for making electrochemical sensors [62-75].

Ionic liquids are the other conductive mediators that can create a synergic effect with nanomaterials for the fabrication of highly sensitive electrochemical sensors [76-80].

In this research, CPE/MBIZLB/CdO-SWCNTs was introduced as a new powerful sensor for the nanomolar determination of sunset yellow in food samples. According to I-V results, the CPE/MBIZLB/CdO-SWCNTs was successfully used for the determination of SY in foods produced with acceptable recovery data.

2. EXPERIMENTAL

2.1. Materials and instruments

Sunset yellow, cadmium acetate dehydrate, carbon nanotube, single-walled/carboxylic acid functionalized, sodium hydroxide (SWCNTs/COOH), 1-methyl-3-butylimidazolium bromide, graphite powder, phosphoric acid, paraffin oil, and hydrochloric acid used in this study, were purchased from Sigma and Merck Company. Stock solution of SY (0.01 M) was prepared by dissolving 0.452 g sunset yellow into 100 mL phosphate buffer solution (PBS) pH=4.0. Thereafter, I-V curves were recorded using a µ-Autolab instrument connected to three electrodes cells. Ag/AgCl/KCl was used as the reference electrode in I-V investigations. CdO-SWCNTs was synthesized using a simple precipitation method reported in a study by Cherghai et al. [81].

2.2. Preparation of CPE/MBIZLB/CdO-SWCNTs

The 0.97 g graphite powder + 0.03 g CdO-SWCNTs was mixed with hand in the presence of 8 drops of paraffin oil as well as 2 drops of MBIZLB for the fabrication of CPE/MBIZLB/CdO-SWCNTs into mortar and pestle. The obtained paste was put at the end of glass tube, and copper wire was then used for electrical conductivity.

2.3. Preparation of real sample

Apple and orange juices were purchased from local market and then centrifuged for 30 min at 300 rpm. The obtained juice was diluted by PBS (pH=4.0) and then used for real sample analysis.

3. RESULTS AND DISCUSSION

3.1. CdO-SWCNTs characterization

The CdO-SWCNTs nanocomposite was characterized by FESEM method. The FESEM image of CdO-SWCNTs nanocomposite clearly confirmed the decoration of CdO nanoparticle at surface of SWCNTs with a good distribution [81].

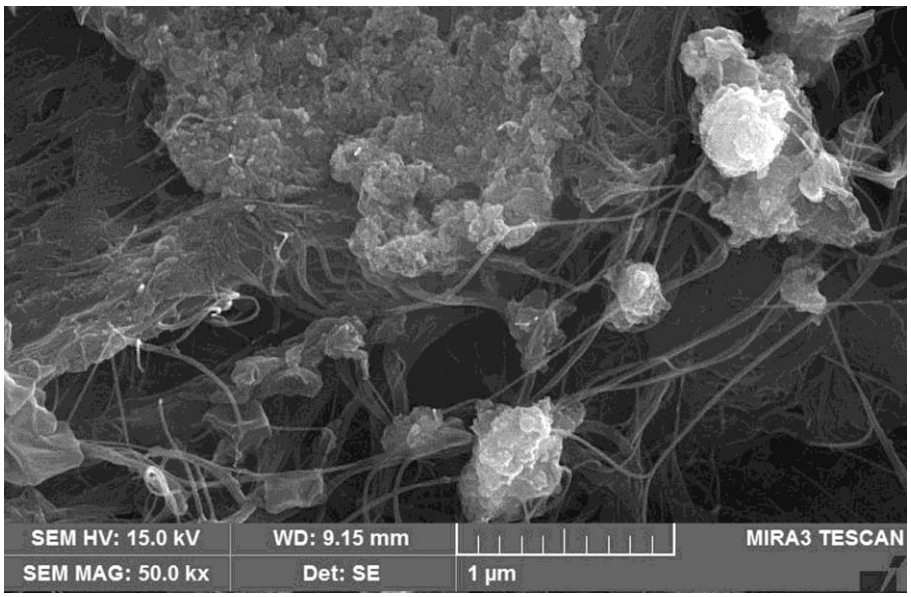


Figure 1. FESEM image of CdO-SWCNTs composite

3.2. Electrochemical behavior

The relationship between oxidation signal of SY and changes in pH was investigated in this step. For this goal, square wave voltammograms (SWV) of 40 µM SY was recorded in the pH range of

3.0 to 5.0 (Figure 2). The results displayed a linear relationship between the potential and pH in this pH range with slope of 58.6 mV/pH that confirmed the suggested mechanism, which is reported in scheme 1 [8]. According to SW voltammograms, the maximum oxidation current for SY was observed at pH=4.0 and this value was selected for next steps.

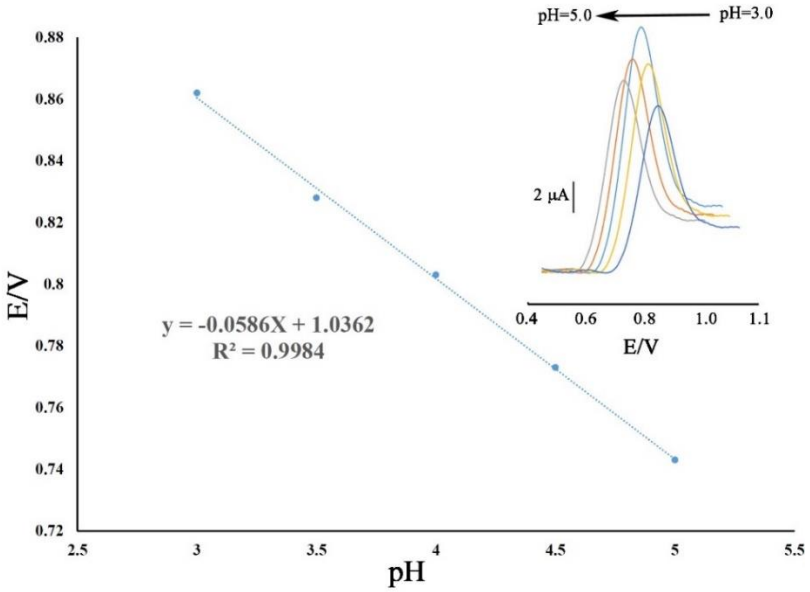
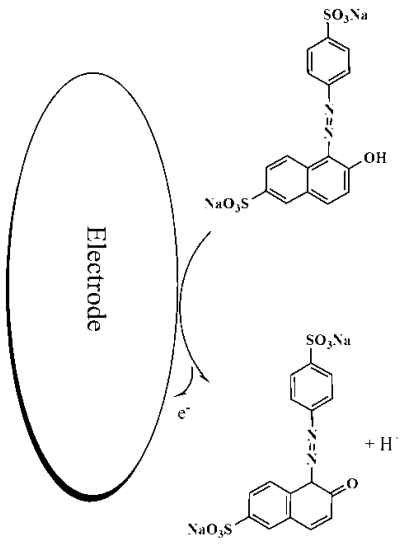


Figure 2. E-pH curve for electro-oxidation of 40 µM SY. Inset) SW voltammograms of 40 µM SY in the different pH range.

The catalytic activity of CPE/MBIZLB/CdO-SWCNTs on oxidation signal of SY was investigated by recording square wave voltammograms of 40 µM SY at the surface of CPE (figure 3 curve a), CPE/CdO-SWCNTs (figure 3 curve b), CPE/MBIZLB (figure 3 curve c), and CPE/MBIZLB/CdO-SWCNTs (figure 3 curve d).



Scheme 1. Sunset yellow electrooxidation mechanism

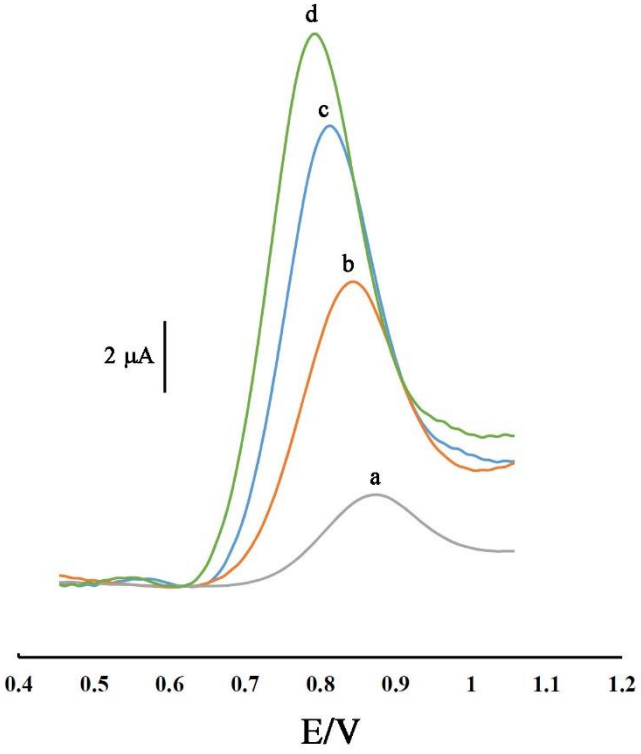


Figure 3. SW voltammogram of 40 µM SY at surface of a) CPE, B) CPE/CdO-SWCNTs, C) CPE/MBIZLB and d) CPE/MBIZLB/CdO-SWCNTs

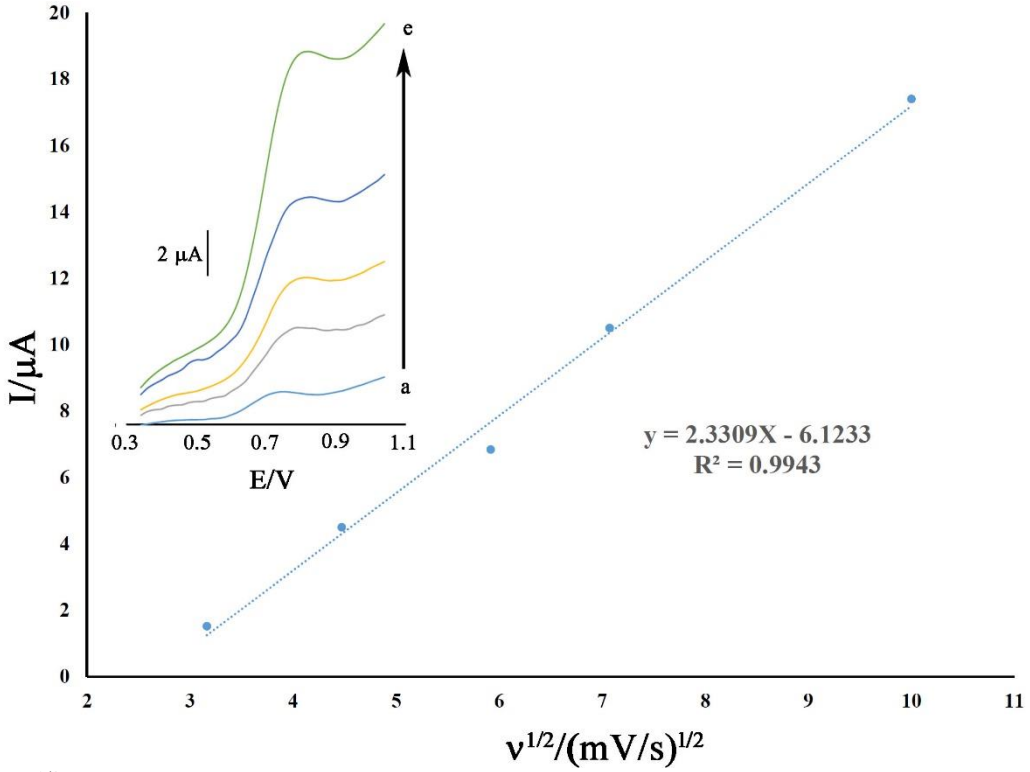


Figure 4. I- $\nu^{1/2}$ curve for electro-oxidation 300 µM SY at scan rates a) 10, b) 20, c)35, d) 50 and e) 100 mV/s.

According to the recorded voltammograms, by moving CPE to CPE/MBIZLB/CdO-SWCNTs, the oxidation current of SY has increased from 2.53 µA to 15.5 µA, confirming the catalytic activities of MBIZLB and CdO-SWCNTs after the modification of PE. This point confirm catalytic activity of nanomaterials and ionic liqouds for modification of sensors [82-87].

In this study, a linear relationship was detected between the oxidation currents of SY vs. $\nu^{1/2}$ using the linear sweep voltammetric method (Figure 4). Accordingly, this relationship confirm diffusion process [88-94] for electro-oxidation of SY at the surface of CPE/MBIZLB/CdO-SWCNTs.

The SW voltammograms of SY and its concentration ($I = 0.2996\ C + 2.7555$; $R^2 = 0.9934$) had a linear relationship in the range of 0.0006 to 180 µM, which was shown using CPE/MBIZLB/CdO-SWCNTs as the working sensor. The CPE/MBIZLB/CdO-SWCNTs showed a detection limit of 0.3 nM for the determination of SY in aqueous solution. The values of LDR and LOD is comparable with previous suggested sensor for determination of SY (Table 1).

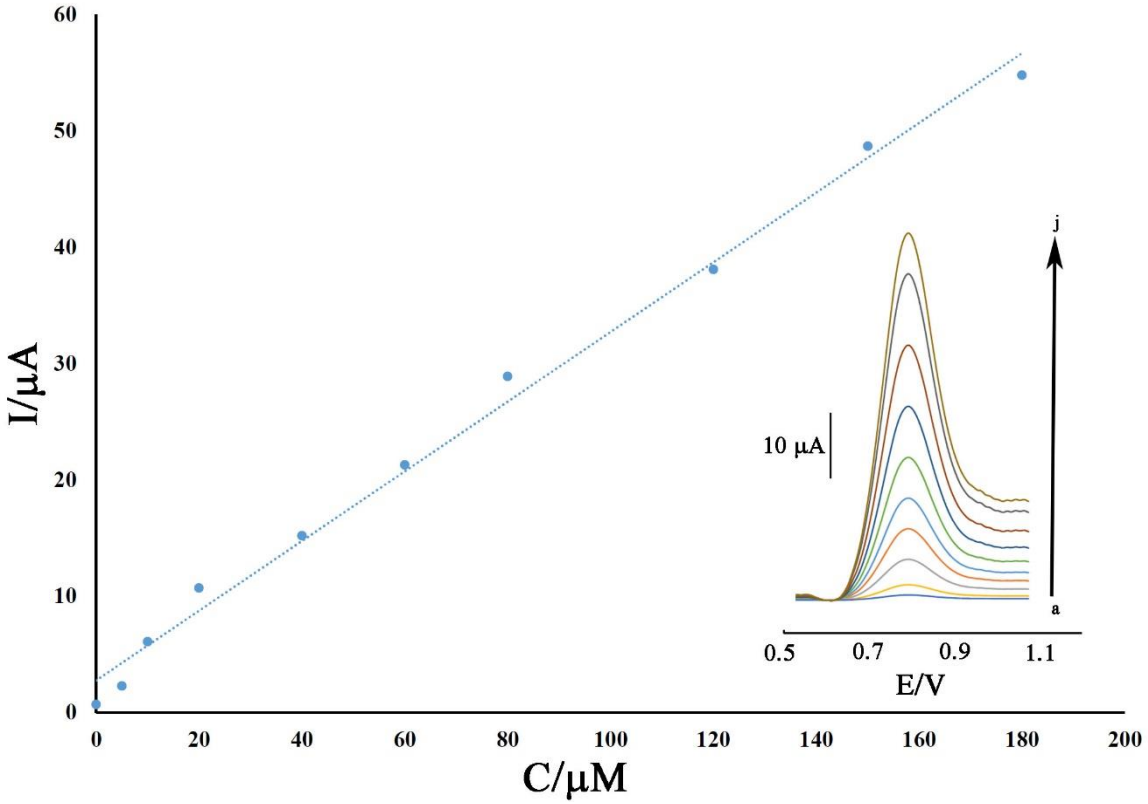


Figure 5. I-C curve for electro-oxidation of SY. Insert) SW voltamograms of SY in the concentration range a) 0.0003; b) 5.0; c) 10.0; d) 20.0; e) 40.0; f) 60.0; g) 80.0; h) 120.0; i) 150 and j) 180 µM.

The interference study showed that CPE/MBIZLB/CdO-SWCNTs could be used as a powerful sensor for the determination of SY in real samples. Regarding the results, the presence of inorganic, organic, and other food additives indicated no important interference for the determination of 10 µM SY at pH=4.0 (Table 2).

Table 1. Analytical data relative to suggested sensors for determination of SY.

Electrode	Mediator	LDR (µM)	LOD (µM)	Ref.
Glassy carbon electrode	Exfoliated graphene oxide	0.05-1.0	0.019	[95]
Au	Poly(aniline-co-o-anisidine-co-o-toluidine)/graphene oxide nanocomposite	5.0-500	0.142	[96]
Graphite Paste Electrode	Attapulgite	0.0025-1.5	0.001	[97]
Glassy carbon electrode	Graphene oxide/multi wall carbon nanotubes composite	0.09-8.0	0.025	[98]
CPE	ZnO-Pd/Ionic liquid	0.001-280	0.0004	[99]
CPE	MBIZLB/CdO-SWCNTs	0.0006-180	0.0003	This work

Table 2. The selectivity test of CPE/MBIZLB/CdO-SWCNTs for determination of 10 µM SY

Species	Tolerant limits ($W_{interference}/W_{SY}$)
Cl^-, Na^+, Li^+, F^-, Ca^{2+}	1000
Alanine and methionine	800
Glucose	500

On the other hand, ability of CPE/MBIZLB/CdO-SWCNTs for the determination of SY was checked using the standard addition method. The results are presented in Table 2 and recovery data confirmed the powerful ability of CPE/MBIZLB/CdO-SWCNTs for the determination of SY in real sample.

Table 3. Determination of SY using CPE/MBIZLB/CdO-SWCNTs in food samples (n=5)

Sample	Exist (µM)	Found (µM)	Recovery%
Orange juices	---	20.33±0.55	---
	10.00	30.89±0.95	101.84
Appel juices	---	15.11±0.45	
	10.00	24.83±1.15	98.88

In the final step, the stability of CPE/MBIZLB/CdO-SWCNTs was checked in a 45-day period. After 45 days, 93% of its initial oxidation current remained that confirmed a good stability of CPE/MBIZLB/CdO-SWCNTs as a new sensor.

4. CONCLUSION

A highly sensitive and simple food electrochemical sensor was fabricated for the determination of SY in food products. The CPE/MBIZLB/CdO-SWCNTs showed a good electro-catalytic activity towards the oxidation of SY at pH=4.0 as an optimum condition. The SW investigation confirmed the ability of CPE/MBIZLB/CdO-SWCNTs for the determination of SY in concentration range of 0.0006 to 180 µM. The CPE/MBIZLB/CdO-SWCNTs showed a powerful ability as a sensor for the determination of SY in food products such as orange and apple juices samples.

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