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Tert-butylhydroquinone Anti-oxidant Voltammetric Sensor as Powerful Tool for Food Sample Analysis

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Abstract- ZnO-CNTs and 1-butyl-3-methylimidazolium bromide (BMIB) ionic liquid were used as amplifiers for modification of carbon paste electrode (CPE) as simple and powerful electrochemical tool for analysis of tert-butylhydroquinone (TBHQ). The pH response on electro-oxidation signal of TBHQ was investigated as effective factor on voltammetric investigation. Under the optimized conditions, a linear dynamic rage 0.09–750 μmol L^{-1} was detected for determination of TBHQ at surface of CPE/BMIB/ZnO-CNTs. The CPE/BMIB/ZnO-CNTs was successfully used as electro-chemical food sensor for determination of TBHQ in food samples.

Keywords- Tert-butylhydroquinone analysis, Voltammetric sensor, ZnO-CNTs nano-composite, Food electrochemical sensor

1. INTRODUCTION

TBHQ is a phenolic antioxidant and a hydroquinone derivative with many applications in different food compounds such as vegetable oils [1-3]. As a food additive, the TBHQ does not change flavor or odor of food compounds and used a useful antioxidant in the recent years [4,5]. On the other hand, TBHQ is a widely used antioxidant in frozen fish and fish products with the highest limit (1 g/kg). Due to wide range, application of TBHQ the fabrication of analytical sensor for determination of this antioxidant is very important in food samples [6-9].

There are different analytical methods such as HPLC, spectroscopy and electrochemical strategy for determination of food additive and especially TBHQ [11-15]. The electrochemical-based methods showed more attention for determination of drugs and food additive due to more advantage compare to other methods in the recent years [16-20]. The electrochemical-based sensors with good ability for the simultaneous determination of electro-active compound and are the best choice for analytical determination of electro-active compounds and especially food additives [21-25]. The low oxidation/reduction signal is the main disadvantage of food compounds for electrochemical analysis that this problem could be resolved by fabrication of voltammetric sensors modified with nano-materials and other electro-active mediators such as ionic liquids [26-28].

Carbon based nanocomposited and especially metal oxide-carbon based nano-composite were suggested as conductive mediators for modification of electrochemical sensors in the recent years [29-35]. The high electrical conductivity of carbon nanotubes and metal oxide nanoparticles could be resolved over-potential redox signal problem for food additive analysis [36].

In this research, we design a highly sensitive electro-analytical tool based on carbon paste electrode amplified with ZnO-CNTs and BMIB as mediators. The CPE/BMIB/ZnO-CNTs was showed a powerful ability as electro-analytical sensor for determination of TBHQ in different food samples.

2. EXPERIMENTAL PART

2.1. Apparatus and materials

All of the voltammetric investigation were performed by a µ-Autolab (µ3AUT 71226) PGSTAT using NOVA software and $Ag/AgCl/KCl_{sat}$ and Pt wire as reference and counter electrodes. The CPE/BMIB/ZnO-CNTs was used as working electrode in electrochemical analysis. The TBHQ, Zinc nitrate and graphite powder were purchased from Sigma-Aldrich. The sodium hydroxide and paraffin oil were purchased from Acros. The ZnO-CNTs was synthesized according to our published paper report procedure [36].

2.2. Preparation of the CPE/BMIB/ZnO-CNTs

For the preparation of CPE/BMIB/ZnO-CNTs; 0.60 g graphite powder +0.40 g ZnO-CNT was mixed and suitable amount paraffin+ BMIB were used as binders for preparation of carbon paste electrode. The obtained paste was packed into end of glass tube and using copper wire the electrical conductivity of sensors was reached.

3. RESULTS AND DISCUSSION

3.1. Characterization of ZnO-CNTs

At first for characterized of ZnO-CNTs, we use transmission electron microscopy (SEM) for morphological investigation (Fig. 1). The SEM images of nano-composite showed a spherical shape of ZnO coated at surface of CNTs tubes.

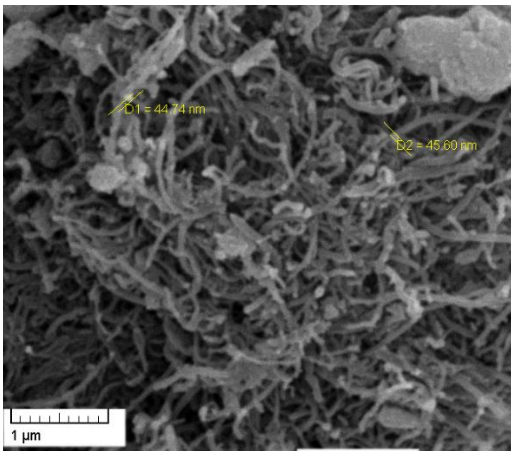
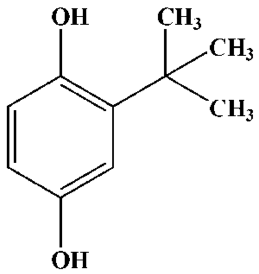


Fig. 1. SEM image of ZnO-CNTs nanocomposite

3.2. Electrochemical Investigation

The phenolic structure of TBHQ (Scheme 1) show that oxidation reaction of this antioxidant could be relative to pH values.



Scheme 1. The chemical structure of TBHQ

The differential pulse voltammograms (DPV) of 350.0 μmol L^{-1} TBHQ were recorded at surface of CPE/BMIB/ZnO-CNTs in the pH ranges of 5.0-9.0 (Fig. 2 inset). As can be seen in figure 2, the maximum oxidation current of TBHQ was occurred at pH=7.0 and this value was selected as optimum condition.

The redox reaction of TBHQ was investigated at surface of CPE/BMIB/ZnO-CNTs (curve a), CPE/BMIB (curve b), CPE/ZnO-CNTs (curve c) and CPE (curve d). With moving CPE to CPE/BMIB/ZnO-CNTs, the oxidation current of TBHQ was increased and oxidation potential of this antioxidant was shifted to negative value that confirm the good electrical conductivity of BMIB and ZnO-CNTs at surface of CPE/BMIB/ZnO-CNTs. The coupling of on BMIB and ZnO-CNTs for modification of CPE showed a good catalytic activity on oxidation signal of TBHQ.

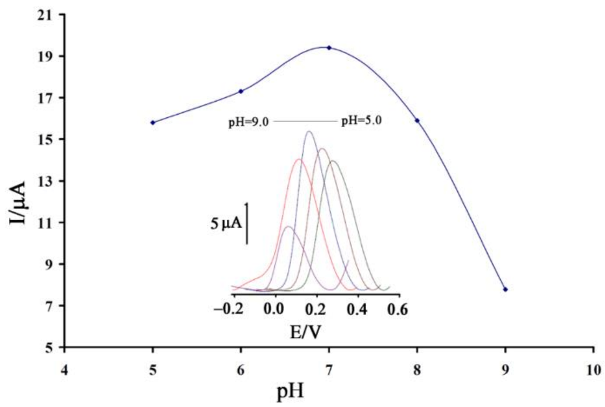


Fig. 2. Current–pH curve for electro-oxidation of 350.0 µmol L^{-1} at surface of CPE/BMIB/ZnO-CNTs. The DPVs of TBHQ recorded at different pH

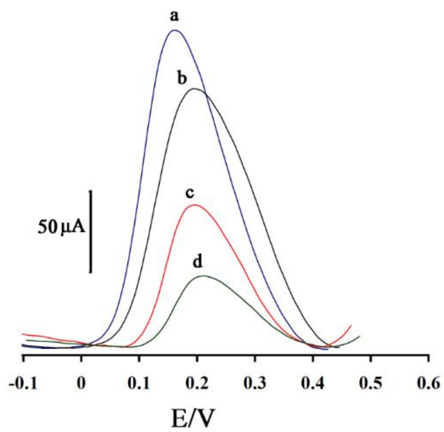


Fig. 3. DPVs of TBHQ at surface of CPE/BMIB/ZnO-CNTs (a); CPE/BMIB (b), CPE/ZnO-CNTs (c) and CPE (d) at a pH=7.0

The redox signal of TBHQ was recorded at different scan rate (Figure 4, inset). A linear relation between oxidation current of TBHQ and $v^{1/2}$ was detected (Figure 4) that confirm a diffusion process [37] for electro-oxidation of TBHQ at surface of CPE/BMIB/ZnO-CNTs.

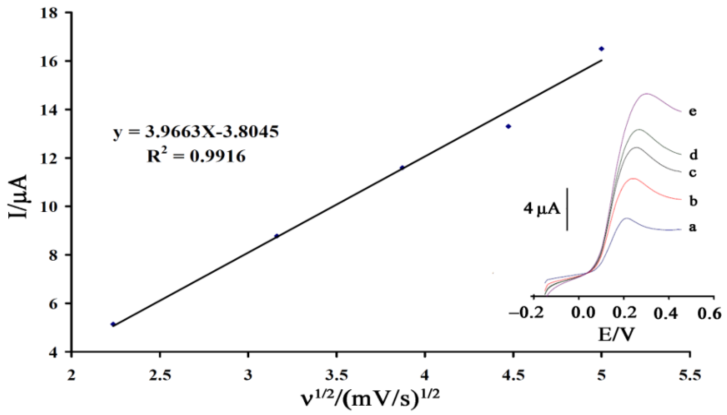


Fig. 4. Plot of current *vs*. $\nu^{1/2}$ for the oxidation of TBHQ at CPE/BMIB/ZnO-CNTs. Inset shows linear sweep voltammograms of TBHQ at CPE/BMIB/ZnO-CNTs at scan rates of a) 5.0, b) 10.0, c) 15.0, d) 20.0, and e) 25.0 mVs^{-1}.

The chronoamperogram of 1.0 mM of TBHQ was recorded at surface of CPE/BMIB/ZnO-CNTs using applied potential 300 mV at pH=7.0 (figure 5). The value of diffusion coefficient (D) of TBHQ was obtained ~7.8×10^{-5} cm^2/s using Cottrell equation and obtained slope from figure 5 inset.

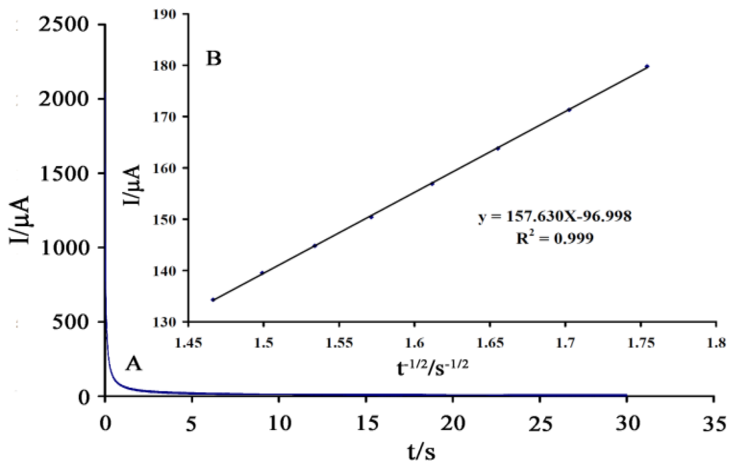


Fig. 5. A) Chronoamperogram obtained for 1.0 mM TBHQ at surface of CPE/BMIB/ZnO-CNTs; B) Cottrell's plot for the data from the chronoamperogram

The DPVs of TBHQ was recorded in the concentration range 0.09–750 μmol L^{-1} at surface of CPE/BMIB/ZnO-CNTs (figure 6 inset). The results confirm a linear relation between TBHQ concentration and oxidation current with a good detection limit of 0.05 μmol L^{-1}.

3.3. Interference Study

The selectivity of the CPE/BMIB/ZnO-CNTs in the analysis of 20.0 μmol L^{-1} TBHQ was investigated in the presence other foreign species. Using acceptable error 5% in current the CPE/BMIB/ZnO-CNTs showed good selectivity for determination of TBHQ at pH=7.0 (see table 1).

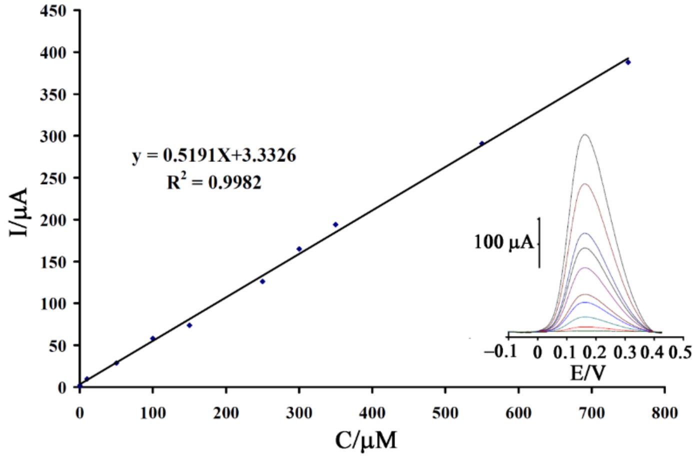


Fig. 6. The current-concentration curve for electro-oxidation of TBHQ at surface of CPE/BMIB/ZnO-CNTs. Inset) DPV of CPE/BMIB/ZnO-CNTs containing different concentrations of TBHQ

Table 1. Interference study for the determination of 20.0 μmol L^{-1} TBHQ

Species	Tolerante limits ($W_{Substance}/W_{Analytes}$)
Na^+, K^+, Li^+, Cl^-, SO_4^{2-}, and F^-	700
Starch	Saturation
glucose, fructose, glycine, lactose, phenylalanine, sucrose	1000

Table 2. Determination of TBHQ in food samples

Sample	Added (μmol L^{-1})	Expected (μmol L^{-1})	Founded (μmol L^{-1})	HPLC method (μmol L^{-1}) [38]	F_{ex}	F_{tab}	t_{ex}	$t_{tab(95\%)}$
Sesame oil	—	—	7.55±0.62	7.87±0.85	5.4	19	1.7	3.8
	10.00	17.55	17.48±0.86	17.22±0.67	6.3	19	0.9	3.8
Soybean oil	—	—	5.50±0.47	5.68±0.83	4.3	19	1.3	3.8
Colza oil	—	—	15.67±0.88	14.98±1.04	9.5	19	1.9	3.8

±Shows the standard deviation (n=3)

3.4. Real food Sample Analysis

For evaluating the analytical performance of CPE/BMIB/ZnO-CNTs in real sample analysis, we use the oil samples as a food real sample for this goal. The results are listed in Table 2 and obtained data confirm high performance ability of CPE/BMIB/ZnO-CNTs for determination of TBHQ as analytical sensor.

4. CONCLUSION

In this project, the CPE/BMIB/ZnO-CNTs was fabricated as highly sensitive analytical tool for electrochemical determination of TBHQ. The ZnO-CNTs was synthesized as conductive mediator and characterized with SEM method. A linear dynamic rage 0.09–750 μmol L^{-1} was detected for determination of TBHQ at surface of CPE/BMIB/ZnO-CNTs and sensor was successfully used for determination of TBHQ in food samples.

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