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Monitoring of Amaranth in Drinking Samples using Voltammetric Amplified Electroanalytical Sensor

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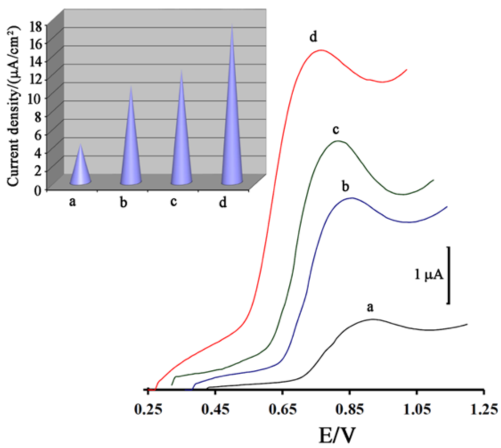
ABSTRACT

Electrochemical sensing of amaranth was performed by a voltammetric strategy in an aqueous solution. The paste electrode (PE) was modified with 1-methyl-3-butyl imidazolium bromide (MBIB) and Pt-CNTs nanocomposite and the MBIB/Pt-CNTs/PE was successfully used as a monitoring tool for sensing of amaranth. The MBIB/Pt-CNTs/PE accelerated the electron transfer in redox reaction of amaranth and improved its oxidation signal by about 4.82 fold. The MBIB/Pt-CNTs/PE measured this azo dye with a detection limit of 1.0 nM. In the final step, MBIB/Pt-CNTs/PE was employed for monitoring of amaranth drinking with an acceptable recovery data.

GRAPHICAL ABSTRACT



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Introduction

One of the most essential issues in advanced cultures is paying attention to human health [1-3]. Meanwhile, food products are the major part of the concerns [4]. Therefore, there are several reports on the quality of food products. Analytical procedures are one of the most important tools for determining the quality of food [5-7]. Chromatography, spectroscopy and electrochemical sensors are among the common methods in assessing the quality of food products [8-10]. In the meantime, electrochemical methods have received more attention than the other approaches due to several advantages, including biocompatibility, and non-use of toxic solvents [11-16]. The potential of electrochemical methods and the variety of their applications have grown significantly due to the use of modifiers such as nanomaterials and ionic liquids [17-23]. Modification of electrochemical sensors with conductive mediators is a viable strategy for improving electrochemical sensors [24-31]. Nanomaterials have created a new perspective in the world of science [32-37]. The superior electrical conductivity of some nanomaterials has created exceptional conditions for the preparation of modified electrodes [38-41].

Azo dyes are an important additive in food, especially beverages [42,43]. These compounds are found in most food products with high color diversity to create attractiveness [44]. Amaranth is an azo dye with red color and employed in different food products [45]. Measuring of amaranth is a good way to track its concentration in food products [46,47]. Therefore, many analytical and especially electrochemical sensors were used for sensing of amaranth [48,49]. As an example, Tajik et al. reported a new voltammetric strategy based on using an graphite electrode amplified with Ni–Mo-MOF nanocomposite for sensing of amaranth with detection limit 0.05 μM in different food samples [50].

This study focused on the fabrication of a powerful and sensitive electroanalytical sensor for determination of amaranth in trace concentrations. The MBIB/Pt-CNTs/PE was introduced as an electroanalytical tool for sensing amaranth, and the results confirmed its capability of sensing this azo dye in different food products. Due to the high conductivity of the mediators, the MBIB/Pt-CNTs/PE sensed amaranth even in a nanomolar concentration with good selectivity. The limit of detection and the linear dynamic range of the proposed sensor is better than other reported electrochemical sensor for monitoring of amaranth in food samples.

Materials and Methods

Amaranth (80 %), graphite powder, SWCNTs, methionine (≥98%), alanine (≥98%), ascorbic acid (99%), vitamin B_2, glucose (≥99%), sodium hydroxide (≥97.0%), 1,2-hexadecanediol, platinum(II) acetylacetonate and paraffin oil were purchased from Sigma-Aldrich Company. The Pt-CNTs nanocomposite was synthesized according to the previously reported strategy by Maleh et al [51].

Instrument

All of electrochemical signals were acquired in a phosphate buffer solution (PBS) through Autolab machine. The Ag/AgCl/KCl_{sta} was employed as a reference electrode in all of experimental. A pH meter type 713 (Metrohm) was utilized in the preparation of PBS.

Preparation of MBIB/Pt-CNTs/PE

The MBIB/Pt-CNTs/PE was prepared by hand-mixing of 0.06 g Pt-CNTs nanocomposite + 0.94 g graphite powder in the presence of 8 drops of paraffin oil + 2 drops MBIB as binder.

Real sample preparation

Orange and apple juices were purchased from local markets, followed by centrifugation and filtering. The pure juice was diluted by PBS (pH=7.0) and used as a real sample for monitoring of amaranth by the standard addition method.

Results and discussion

Optimization of electrode fabrication

For fabrication of MBIB/Pt-CNTs/PE, the ratio of MBIB to paraffin oil and Pt-CNTs nanocomposite to graphite powder was optimized in the solution containing 500 μM amaranth. As can be seen in Figure 1, with the addition of Pt-CNTs nanocomposite in the carbon paste matrix, the conductivity of the electrode increased. At a ratio of 6% (w:w) Pt-CNTs nanocomposite to graphite powder, the maximum conductivity detected and this value of nanocomposite was employed for fabrication of MBIB/Pt-CNTs/PE.

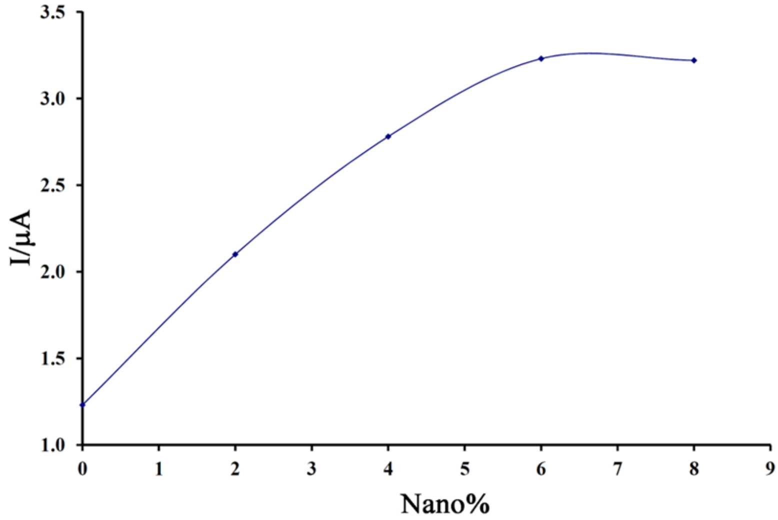

Figure 1: I vs. nanocomposite percentage curve for electro-oxidation of amaranth

On the other hand, the replacement of MBIB instead of paraffin in the carbon paste matrix improved the oxidation signal of amaranth (Figure 2). The maximum conductivity was detected in the presence of 20 (v:v) of MBIB in the electrode matrix. The MBIB/Pt-CNTs/PE was fabricated utilizing optimized data and employed in the following steps, as described in section 2.3.

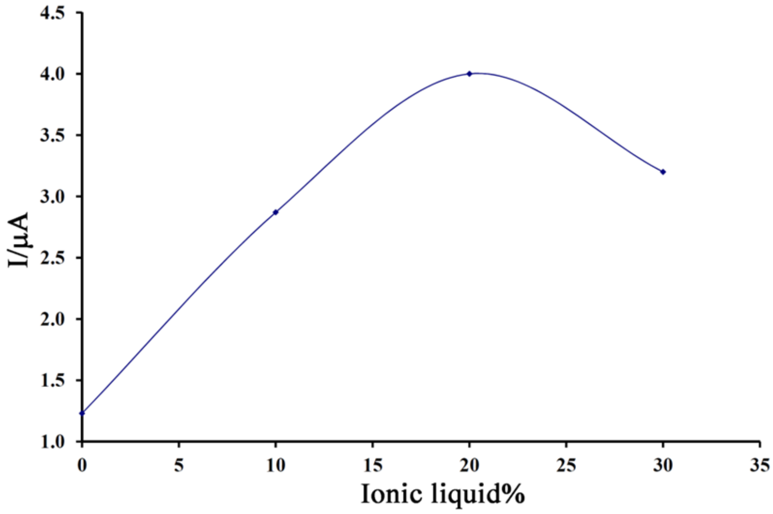

Figure 2: I vs. MBIB percentage curve for electro-oxidation of amaranth

pH investigation

The oxidation signal of amaranth was recorded in pH range of 4.0-8.0 by linear sweep voltammetry method (LSV) (Figure 3 inset). A negative shift was observed in the recorded signals that confirmed the presence of proton in redox reaction of amaranth. Linear relation between potential and pH with slope -0.052 V/pH confirmed the equal value of electron and proton took place in oxidation/reduction reaction of amaranth.

Catalytic effect

The LS voltammograms of 500 µM amaranth were recorded at the surface of PE (curve a), Pt-CNTs/PE (curve b), MBIB/PE (curve c), and MBIB/Pt-CNTs/PE (curve d), respectively (Figure 4).

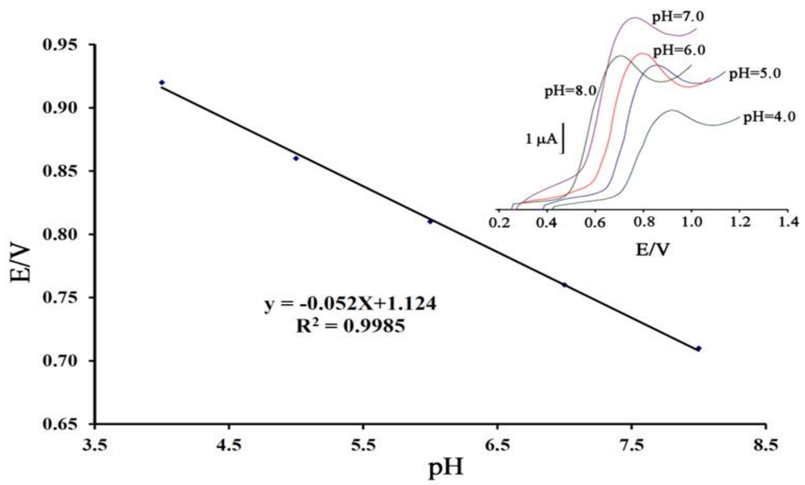

Figure 3: E-pH curve for electrooxidation of 500 μM amaranth at surface MBIB/Pt-CNTs/PE in the different pH. Inset) Relative LS voltammograms

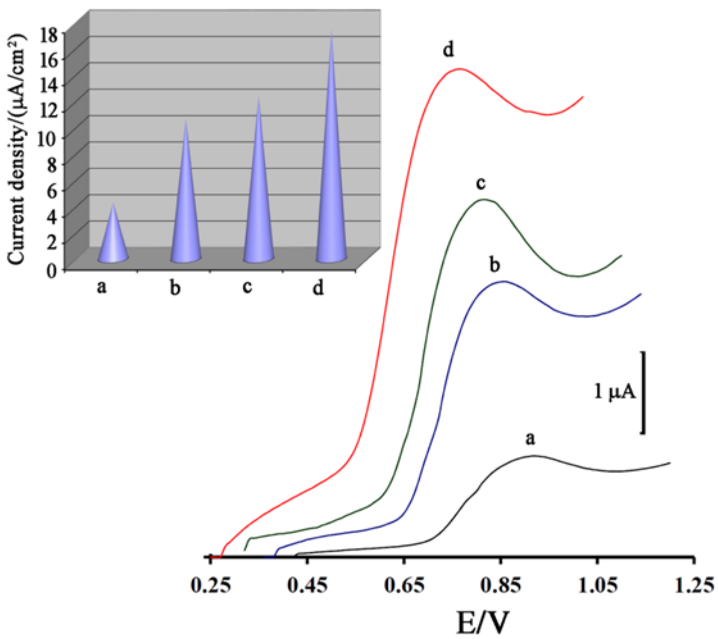

Figure 4: LSV 500 μM amaranth at surface PE (a), Pt-CNTs/PE (b), MBIB/PE (c) and MBIB/Pt-CNTs/PE (d)

As can be seen, the current of amaranth was improved from 1.23 μA to 5.94 μA with modification of PE with MBIB and Pt-CNTs, revealing the high conductivity of MBIB and Pt-CNTs as a new catalysts.

Analytical parameters

Analytical parameters such as LOD and LDR were computed for monitoring of amaranth at the surface of MBIB/Pt-CNTs/PE using differential pulse voltammetric method (not shown). A linear relation with an equation of I = 0.1052 C + 2.4784 (r^2 = 0.9939) was detected for monitoring of amaranth in the concentration range of 0.005 – 310.0 μM. The MBIB/Pt-CNTs/PE showed a detection limit of 1.0 nM towards sensing of amaranth at the optimum conditions.

Determination of amaranth in real samples

The MBIB/Pt-CNTs/PE was utilized for monitoring of amaranth in the orange and apple juices by standard addition methods. The results were tabulated in Table 1 and the recovery data was clearly revealed the high capability of MBIB/Pt-CNTs/PE toward sensing and monitoring of amaranth.

Table 1: Determination of amaranth in drinking samples

Sample	Amaranth added (μM)	Amaranth expected(μM)	Amaranth founded (μM)	Recovery%
Orange juice	---	---	<LOD	---
	10.00	10.00	9.83±0.43	98.3
	---	---	<LOD	---
	10.00	10.00	10.41±0.57	104.1

Conclusion

In this research work, the MBIB/Pt-CNTs/PE was introduced as a new electroanalytical sensor for monitoring of amaranth. The MBIB/Pt-CNTs/PE offered an high catalytic activity of oxidation signal of amaranth and detected this azo dye in a concentration range of 0.005 – 310.0 μM. The MBIB/Pt-CNTs/PE was successfully empolyed for sensing of amaranth in the drinking juice samples with a recovery range of 98.3% - 104.1%.

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Authors' contributions

All authors contributed toward data analysis, drafting and revising the paper and agreed to responsible for all the aspects of this work.

Conflict of Interest

We have no conflicts of interest to disclose.

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