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Analytical Methods

A new strategy for determination of bisphenol A in the presence of Sudan I using a ZnO/CNTs/ionic liquid paste electrode in food samples

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ABSTRACT

The electrochemistry of bisphenol A (BPA) was studied by voltammetric methods at a surface of carbon paste electrode modified by a ZnO/CNTs nanocomposite and room-temperature ionic liquid of 1,3-dipropylimidazolium bromide. The ratio of ZnO/CNTs and ionic liquid (IL) on the surface of the electrode has to be controlled carefully because the charging currents. The anodic peaks of BPA and Sudan I in their mixture can be well separated. At pH 7.0 the two peaks are separated ca. 0.47 and 0.70 V, respectively; hence BPA can be determined in the presence of Sudan I and more than 8.7 times current excess of BPA. The peaks current of square wave voltammograms (SWV) of BPA and Sudan I increased linearly with their concentration in the ranges of 0.002–700 μmol L^{-1} BPA and 0.2–800 μmol L^{-1} Sudan I. The detection limits for BPA and Sudan I were 9.0 nmol L^{-1} and 80 nmol L^{-1}, respectively. The modified electrode has been successfully applied for the assay of BPA in food samples. This study provides a simple and easy approach to selectively detect BPA in the presence of Sudan I.

1. Introduction

Foodborne illnesses are a burden on public health and contribute significantly to the cost of health care. Food safety is a scientific discipline describing handling, preparation, and storage of food in ways that prevent foodborne illness. This includes a number of routines that should be followed to avoid potentially severe health hazards (http://en.wikipedia.org/wiki/Food_safety; 30 December, 2013). Bisphenol A is a chemical used in the manufacture of epoxy resins for the lacquer lining of metal food cans, as a monomer of polycarbonates, and the monomer of plastic used in the base paste of dental sealants (Zhuang, Zhang, & Cao, 2008). But polycarbonate and epoxy resins, inner surface coating of food and beverage cans, from which BPA can leach into food and environment (Kim, Kim, Kim, Choi, & Lee, 2013). Exposure to BPA has resulted to extensive human health effects since it exhibits estrogenic activity. These include reproduction dysfunctions, endometrial hyperplasia, recurrent miscarriages, abnormal karyotypes and polycystic ovarian syndrome. Therefore, it is quite urgent to search for an efficient approach for the degradation and detection of such a chemical so as to minimize its contamination in food samples. On the other hand, because of its ubiquitous nature and its endocrine disrupting potential, BPA has been included in environmental water monitoring or determination studies using several methods (Ntsendwana, Mamba, Sampath, & Arotiba, 2012). Between all of the methods that used for determination of BPA, electrochemical methods have attracted more attention in two decades for due to their sensitivity, accuracy; lower cost, high dynamic range and simplicity (Sanghavi & Srivastava, 2011; Gupta, Atar, Yola, Üstündağ, & Uzun, 2014; Sanghavi et al., 2013; Yola, Atar, Qureshi, Üstündağ, & Solak, 2012; Yola, Atar, Üstündağ, & Solak, 2013; Sanghavi & Srivastava, 2010; Gupta, Singh, Al Khayat, & Gupta, 2007; Goyal, Gupta, & Chatterjee, 2009; Gupta, Singh, Ashok, Mehtab, & Gupta, 2006; Gupta, Jain, Maheshwari, Lang, & Ishtaiwi, 2006; Gupta et al., 2011). Therefore, electrochemical methods were used as performance techniques for determination of some pharmaceutical, biological, environmental and food compounds in recent years (Gupta, Chandra, & Lang, 2005; Gupta, Prasad, & Kumar, 2003; Gupta, Jain, & Kumar, 2006; Sanghavi et al., 2013; Sanghavi & Srivastava, 2013; Goyal, Gupta, & Bachheti, 2007; Gupta et al., 2014; Raoof, Ojani, & Karimi-Maleh, 2008; Yola & Atar, 2014; Yola, Gupta, Eren, Şen, & Atar, 2014; Yola et al., 2014; Gupta, Singh, & Gupta, 2007; Gupta, Chandra, & Mangla, 2002; Gupta, Jain, & Khurana, 1997; Gupta, Agarwal, & Singhal, 2011; Gupta, Mangla, Khurana, & Kumar, 1999; Gupta, Jain, Kumar, Agarwal, & Maheshwari, 2006).

Sudan I (sudan red), a synthetic azo dye, is considered to be a genotoxic carcinogen and classified as a category 3 carcinogen by the International Agency for Research on Cancer (IARC) (1975)

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(Stiborova, Martinek, Rydlova, Hodek, & Frei, 2002). Unluckily, it was still found in foodstuffs as additive due to its bright colour, low cost, and long maintenance, such as, poultry feed, paprika, ketchup, sausage, pie, etc. So, it is essential to develop a sensitive, selective, simple, fast and accurate method for identification and quantification of Sudan I in foodstuffs (Yin et al., 2011). Sudan I and BPA are two important compound that put in black list as a unsafe compounds in food samples by the International Agency for Research on Cancer (IARC). So, simultaneous determinations of these compounds are very important for customer for prevention of foodborne illnesses.

There are well-developed methods to synthesize nanomaterials such as carbon nanotubes, nanorod, nanocube, nanoshell, metal nanocomposite, which have found real applications in pharmaceutical, food industrial, environmental and biological process (Elyasi, Khalilzadeh, & Karimi-Maleh, 2013; Olivé-Monllau, Martínez-Cisneros, Bartrolí, Baeza, & Céspedes, 2011; Olivé-Monllau, Pereira, Bartrolí, Baeza, & Céspedes, 2010; Olivé-Monllau, Esplandiu, Bartrolí, Baeza, & Céspedes, 2010; Sanghavi, Kalambate, Karna, & Srivastava, 2014). One of the most important functions of nanocomposite is electrocatalysis, especially with noble metal nanoparticles, which have high catalytic activity for many chemical reactions and high surface area for increasing current density (Karimi-Maleh, Biparva, & Hatami, 2013; Moradi et al., 2013; Tavana et al., 2012; Sanati, Karimi-Maleh, Badiei, Biparva, & Ensafi, 2013; Roodbari Shahmiri, Bahari, Karimi-Maleh, Hosseinzadeh, & Mirnia, 2013). Because nanomaterials also have good biocompatibility, they are used for the fabrication of biosensors in biological and food compound analysis (Beitollah et al., 2012; El-Ansary & Faddah, 2010; Ensafi, Karimi-Maleh, Mallakpour, & Hatami, 2011; Ensafi, Karimi-Maleh, Mallakpour, & Rezaei, 2011; Gupta, Jain, Radhapyari, Jadon, & Agarwal, 2011; Wang et al., 2013).

ILs base electrochemical sensors have received much attention due to their specific characteristics such as good chemical and thermal stability, negligible vapour pressure, high ionic conductivity and wide electrochemical windows in the recent years (Afsharmanesh, Karimi-Maleh, Pahlavan, & Vahedi, 2013; Bijad, Karimi-Maleh, & Khalilzadeh, 2013; Ensafi, Bahrami, Rezaei, & Karimi-Maleh, 2013a; Ensafi, Izadi, & Karimi-Maleh, 2013b). ILs can be used to make different modified electrodes, which could increase the transfer rate of different compounds (Sadeghi, Karimi-Maleh, Bahari, & Taghavi, 2013; Salmanpour et al., 2012; Vahedi et al., 2013).

According to the above points, in this study we describe synthesis and application of novel ZnO/CNTs nanocomposite as a novel nanosensor and 1,3-dipropylimidazolium bromide as suitable binder in carbon paste matrix for voltammetric determination of BPA. We have also evaluated the analytical performance of the modified electrode for quantification of BPA in the presence of Sudan I in food samples.

2. Experimental

2.1. Chemicals

All chemicals used were of analytical reagent grade purchased from Merck (Darmstadt, Germany) unless otherwise stated. Doubly distilled water was used throughout.

Bisphenol A (from Merck) stock solution, 1×10^{-3} mol L^{-1}, was prepared by dissolving 0.023 g of the reagent in a 100-mL volumetric flask.

Sudan I (from Fluka) stock solution, 1×10^{-3} mol L^{-1}, was prepared by dissolving 0.028 g of the reagent in a 100-mL volumetric flask (ethanol–water (1:1) solution). Phosphate buffer (sodium dihydrogen phosphate and disodium monohydrogen phosphate plus sodium hydroxide, 0.1 mol L^{-1}) solutions (PBS) with different pH values were used.

High viscosity paraffin (d = 0.88 kg L^{-1}) from Merck was used as the pasting liquid for the preparation of the carbon paste electrodes.

2.2. Apparatus

Cyclic voltammetry, chronoamperometry, and square wave voltammetry were performed in an analytical system, μ-Autolab with PGSTAT (Eco Chemie, the Netherlands). The system was run on a PC using NOVA software. A conventional three-electrode cell assembly consisting of a platinum wire as an auxiliary electrode and an $Ag/AgCl/KCl_{sat}$ electrode as a reference electrode was used. The working electrode was either an unmodified carbon paste electrode (CPE), ZnO/CNTs/IL/CPE, IL/CPE or a ZnO/CNTs/CPE. X-ray powder diffraction studies were carried out using a STOE diffractometer with Cu-Ka radiation (k = 1.54 Å). Samples for transmission electron microscopy (TEM) analysis were prepared by evaporating a hexane solution of dispersed particles on amorphous carbon coated copper grids.

2.3. Synthesis of ZnO/CNTs

The commercial functional multi-walled carbon nanotubes (COOH group) with tube diameters of about 5–10 nm were used. The preparation of ZnO/CNTs catalysts includes two steps. In the first step, certain amounts of purified CNTs (3 g) were dispersed into distilled water solution of NaOH (0.5 M; 50 ml) by ultrasonication for 30 min. The second step is the supporting of zinc oxide on functional carbon nanotubes by a direct deposition process. 3.2 g ZnO $(NO_3)_2{\cdot}6H_2O$ was dissolved in 100 cm^3 distilled water. In the constant magnetic stirring, the solution of ZnO $(NO_3)_2{\cdot}6H_2O$ was added drop wise to the solution of CNTs at 50 °C through a dropping funnel. The rate of addition of the salt solution was kept approximately at 30 ml/h. After completion of the precipitation procedure, the mixture was stirred at room temperature for 12 h, washed and filtered continually in distilled water (pH 7.0), and dried at 120 °C. The solid samples were then calcined at 300 °C for 2 h.

2.4. Preparation of the sensor

ZnO/CNTs/CPE was prepared by hand-mixing of 0.85 g of graphite powder and 0.15 g ZnO/CNTs plus paraffin at a ratio of 70/30 (w/w) and mixed well for 50 min until a uniformly wetted paste was obtained. The paste was then packed into a glass tube. Electrical contact was made by pushing a copper wire down the glass tube into the back of the mixture. When necessary, a new surface was obtained by pushing an excess of the paste out of the tube and polishing it on a weighing paper ZnO/CNTs/IL/CPE was prepared by mixing of 0.2 g of 1,3-dipropylimidazolium bromide, 0.8 g of the liquid paraffin, 0.15 g of ZnO/CNTs, and 0.85 g of graphite powder. Then the mixture was mixed well for 40 min until a uniformly wetted paste was obtained. A portion of the paste was filled firmly into one glass tube as described above to prepare ZnO/CNTs/IL/CPE.

2.5. Preparation of real samples

Fifteen cans of each group of foods (corn, tomato paste, stew, water bottle and tuna fish), totally 45 samples all bearing the same batch number and near their expiration date, were purchased from retail outlets in Sari. The distribution of types of samples tested in this survey was similar across Iran. Purchasing of the samples was carried out in October 2013. Samples were stored and sealed at room temperature. After opening the cans, the total contents of