



# Based on magnetic graphene oxide highly sensitive and selective imprinted sensor for determination of sunset yellow



Jianbo Li, Xiaojiao Wang, Huimin Duan, Yanhui Wang, Yanan Bu, Chuannan Luo\*

Key Laboratory of Chemical Sensing & Analysis in Universities of Shandong (University of Jinan), School of Chemistry and Chemical Engineering, University of Jinan, Jinan 250022, China

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## ABSTRACT

A new imprinted material based on  $\beta$ -cyclodextrin/ionic liquid/gold nanoparticles functionalized magnetic graphene oxide has been successfully synthesized and modified to the glassy carbon electrode surface to construct imprinted electrochemical sensor to detect sunset yellow. The sensitivity and electrochemical response of the electrode can be improved by nanomaterials. The surface morphology and crystal structure of the hybrid nanomaterial has been characterized by scanning electron microscopy, X-ray diffraction, Fourier transform infrared spectroscopy. The electrochemical behaviors of the hybrid nanomaterials based sensor were evaluated through cyclic voltammetry and electrochemical impedance spectroscopy. Under the optimized conditions, the proposed electrochemical sensor showed a fast re-binding dynamics, which was successfully applied to sunset yellow detection with a wide linear range from  $5.0 \times 10^{-9}$  to  $2.0 \times 10^{-6}$  mol L<sup>-1</sup> and a detection limit of  $2.0 \times 10^{-9}$  mol L<sup>-1</sup>. The electrochemical sensor has been successfully applied in the determination of SY in spiked water samples, mirinda drink and minute maid, and the recoveries for the standards added are 97–105%.

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## 1. Introduction

A kind of azo dye-sunset yellow (SY) which is usually added in foods such as beverages, candies, cream products and spicy snacks [1,2]. It can not only change the exterior and texture of food but also maintain the self-color of the food during processing and storage [1,3,4]. However, SY is reported to be attributable to hepatocellular damage and renal failure [5]. The excessive intake of SY can cause allergies, diarrhea and other symptoms [6,7]. Currently, SY is added to a large number of food products, such as cheese, candy, hot strip, pepsi-cola, mirinda soft drinks. Therefore, in order to ensure human health, it is crucial to monitor SY content in food products and looking for a new sensing method to simple, sensitive, economic and rapid detection of SY is very important (Fig. 1).

So far, many methods are mentioned for the detection of SY, including electrochemical method [7], enzyme linked immunosorbent assay [8], high performance liquid chromatography [9], and spectrophotometry [10]. In these methods, the electrochemical method has recently become more research value and practicability. A multitude of electrochemical methods have been reported for detection of SY [7,11,12]. These electrochemical sensor methods were quite sensitive, but the measuring ranges were

narrow and their selectivity was not enough good. To improve the selectivity of electrochemical methods, molecular imprinting technology [13] has been introduced in the field of electrochemistry [14,15]. Molecularly imprinted polymers have some specific advantages, for instance, great specific recognition ability, which is almost not affected by external environment [16], low cost of preparation, high stability, high surface-to-volume ratio and good selectivity [17,18]. However, molecularly imprinted polymers have some disadvantages such as low density of imprinted sites, slow binding time, incomplete template removal and weak electrical conductivity, which limit its application in the field of sensors [19–22]. Fortunately, many new class multifunctional nanomaterials are used to change these disadvantages [23]. Multifunctional nanomaterials molecularly imprinted electrochemical sensors were not only high sensitivity but also great selectivity.

Graphene oxide (GO) a single-atom-thick and two-dimensional carbon material that has attracted great attention because of its remarkable electronic, mechanical, and thermal properties [24,25]. Graphene oxide has good water solubility and its surface has many active sites that can be loaded with nanoparticles. With so many merits, GO has become a widely used material in the field of electrochemical sensors [26–27]. In order to have a better separation and purification of the nanomaterials in the process of preparation, grafted nano Fe<sub>3</sub>O<sub>4</sub> on the surface of GO then magnetic graphene oxide (MGO) was prepared [28].  $\beta$ -Cyclodextrin ( $\beta$ -CD) with a hydrophilic exterior surface and lipophilic cavity [29]

\* Corresponding author.

E-mail address: [chm\\_lijianbo@yeah.net](mailto:chm_lijianbo@yeah.net) (C. Luo).

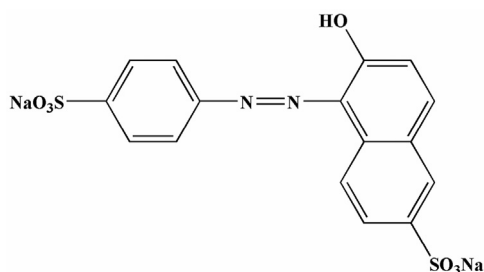


Fig. 1. The molecular structure of sunset yellow.

has strong adsorption capacity, good biocompatibility and can improve the electrochemical response [30].  $\beta$ -CD and GO are combined to form supramolecular systems by non-covalent bonds and dehydration condensation of hydroxyl and carboxyl [31]. The supramolecular systems can improve graphene solubility and prevented from gathering while the two-dimensional material graphene to build a three-dimensional. Hence, there are a wide range of applications in electrochemical sensor. Ionic liquids (IL), a new class of conductive materials made of molten organic cations and various anions [32]. Due to its unique physical properties of wide electrochemical windows, commendable chemical and thermal stability, high ionic conductivity and low toxicity, it can be used not only as the supporting electrolyte but also as the modifier in chemically modified electrode [33]. Gold nanoparticles (AuNPs) with large surface area, high conductivity and electro-catalysis characteristics, have been used to improve the detection limits in electrochemical studies [31,34,35].

In this work, a new and efficient molecularly imprinted nanomaterial based on  $\beta$ -cyclodextrin/ionic liquid/gold nanoparticles functionalized magnetic graphene oxide (MGO/ $\beta$ -CD/IL/AuNPs) has been synthesized and was used to modify glassy carbon electrode surface to construct SY molecularly imprinted electrochemical sensor. Firstly, MGO were synthesized by coprecipitation  $\text{Fe}_3\text{O}_4$  nanoparticles in the surface of GO. Secondly  $\beta$ -CD and IL were adsorbed on MGO by hydrogen bond and electrostatic interaction respectively. Then, AuNPs were assembled on the surface of MGO/ $\beta$ -CD/IL by Au-S bond. Finally, molecularly imprinted polymers (MIPs) were prepared by precipitation polymerization. The electrochemical sensor combines the high selectivity of MIPs and the ultra-sensitivity of multifunctional nanomaterials. Owing to their synergic effect, the constructed electrochemical sensor exhibits excellent selectivity, high sensitivity and fast response towards SY and the sensor has been applied to the detection of SY in actual samples.

## 2. Experimental

### 2.1. Materials and reagents

Sunset yellow, 1-butyl-3-methyl imidazole hydrobromide (purity: 99%), methacrylic acid (MAA) and ethylene glycol dimethacrylate (EDMA) were purchased from Aladdin Chemistry Co. Ltd. (Shanghai, China). Natural graphite powder, anhydrous ethanol, toluene were supplied by Sinopharm Chemical Reagent Co. Ltd. (China). Nitric acid ( $\text{HNO}_3$ ), hydrochloric acid (HCl), sulfuric acid ( $\text{H}_2\text{SO}_4$ ) and acetic acid were purchased from Kant Chemical Co. Ltd. (Laiyang, China). Hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) and chloroauric acid ( $\text{HAuCl}_4$ ) were obtained by the Chemical Reagent Third Company (Tianjin, China). Azodiisobutyronitrile (AIBN) was purchased from Chemical Plant (Beijing, China). All chemicals used in the experimental are of analytical reagent grade. The water in the experimental process used was ultrapure water.

### 2.2. Apparatus

All electrochemical measurements were performed on a CHI 660D electrochemistry workstation (Beijing Huake Putian Technology Co., Ltd. China). The classical three-electrode system was adopted. The working electrode was a glassy carbon electrode modified with molecularly imprinted polymers. The auxiliary and reference electrodes were a Platinum electrode and a saturated calomel electrode (SCE), respectively. Nanophase materials were characterized by Scanning Electron Microscopy (SEM, Japan), X-ray diffraction (XRD, Germany) and Fourier Transform Infrared Spectroscopy (FT-IR). Electrochemical impedance spectroscopy (EIS) was performed on a CHI 604D Electrochemical Workstation (Shanghai CH Instruments Inc., China). SEM observations were carried out using a JEOL JSM-6300 apparatus with an applied voltage of 20 kV. XRD data were collected at room temperature on a Focus D8 Brooke diffractometer with a graphite-filtered  $\text{Cu-K}\alpha$  source ( $\lambda = 0.15418 \text{ nm}$ ), 40 kV, 30 mA. FT-IR spectrum was recorded at room temperature in attenuated total reflection mode (ATR-FTIR) in the wave number range of  $400\text{--}4000 \text{ cm}^{-1}$  on a Nicolet 400 Madison Fourier transform spectrometer. All the electrochemical experiments were carried out at room temperature.

### 2.3. Preparation of imprinted material

#### 2.3.1. Synthesis of GO and MGO

The synthesis method of GO is a modified Hummers method [36–38]. Specific steps: 1.0 g of graphite power and 200.0 mL  $\text{H}_2\text{SO}_4/\text{HNO}_3$  solution ( $v:v=9:1$ ) were mixed and stirring for 0.5 h. Then 6.0 g  $\text{KMnO}_4$  was slowly added into above solution in the condition of ice bath. After stirring for 2 h, transferred to the oil bath pot, stirring for 12 h under the condition of  $90^\circ\text{C}$ , then cooling to room temperature and adding 150 mL  $\text{H}_2\text{O}_2$  solution (30%) keep stirring under the condition of the ice bath. The mixture was separated by centrifugation. The precipitate was washed with HCl solution (0.2 M) and anhydrous ethanol several times until the supernatant pH to 7.0. The last mixture was dried in a vacuum drying box under the condition of  $50^\circ\text{C}$  and GO was obtained.

MGO was prepared by following method [28]. Under  $\text{N}_2$  atmosphere, 50.0 mL solutions containing 50.0 mg GO, 50.0 mg  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$  and 35.0 mg  $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$  was gradually added to  $\text{NH}_3 \cdot \text{H}_2\text{O}$  (28%) to adjust the pH to 9.0, heating and stirring for 30 min under the condition of  $90^\circ\text{C}$ . Prepared MGO was isolated and purified by the magnetic field.

#### 2.3.2. Synthesis of MGO/ $\beta$ -CD and MGO/ $\beta$ -CD/IL

10.0 mg MGO and 40.0 mg  $\beta$ -CD were added to 40 mL ultrapure water. The solution was vigorously shaken 5 min before adds 0.15 mL ammonia and 10.0  $\mu\text{L}$  hydrazine and then transferred to the oil bath pot and heated for 4 hours under the conditions of  $60^\circ\text{C}$ . The MGO/ $\beta$ -CD was precipitated and separated by a magnetic field. 10.0 mg resultant material was added to 150.0 mL anhydrous ethanol and 10.0 mg ionic liquid (1-Butyl-3-methylimidazolium Bromide) sonicated for 30 min. Separation and drying to obtain the MGO/ $\beta$ -CD/IL complexes.

#### 2.3.3. Synthesis of AuNPs and MGO/ $\beta$ -CD/IL/AuNPs

Firstly, the gold nanoparticles were prepared using sodium citrate reduction method of chloroauric acid [39]. 3.65 mL chloroauric acid solution (0.01 M) was diluted into 150 mL and heated to micro-boiling. Then, 3.0 mL sodium citrate was quickly added and keep heating and stirring. When the solution turned wine red to stop heating and keep stirring until the solution was cooled to room temperature. The gold nanoparticle was stored in a

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