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Using nanostructured conductive carbon tape modified with bismuth as the disposable working electrode for stripping analysis in paper-based analytical devices



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ABSTRACT

Low cost disposable working electrodes are specifically desired for practical applications of electrochemical detection considering maturity of electrochemical stations and data collection protocols. In this paper double-sided conductive adhesive carbon tape with nanostructure was applied to fabricate disposable working electrodes. Being supported by indium tin oxide glass, the prepared carbon tape electrodes were coated with bismuth film for stripping analysis of heavy metal ions. By integrating the bismuth modified electrodes with paper-based analytical devices, we were able to differentiate Zn, Cd and Pb ions with the sample volume of around 15 μ L. After the optimization of parameters, including modification of bismuth film and the area of the electrodes, etc., Pb ions could be measured in the linear range from 10 to 500 μ g/L with the detection limit of 2 μ g/L. Our experimental results revealed that the disposable modified electrodes could be used to quantify migrated lead from toys with the results agreed well with that using atomic absorption spectrometry. Although bismuth modification and stripping analysis could be influenced by the low conductivity of the carbon tape, the low cost disposable carbon tape electrodes take the advantages of large-scaled produced double-sided carbon tape, including its reproducible nanostructure and scaled-up fabrication process. In addition, the preparation of disposable electrodes avoids time-consuming pretreatment and experienced operation. This study implied that the carbon tape might be an alternative candidate for practical applications of electrochemical detection.

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1. Introduction

In electrochemical detection, the working electrode plays a pivotal role because all the electrochemical reactions occur on its surface. There have been numerous studies focusing on the working electrode surface, such as chemical modification electrodes [1,2]. Because of the electrochemical reactions, the surface of working electrodes is delicate and could be easily polluted during electrochemical detection. Such contamination would inevitably make the electrochemical detection unstable and irreproducible.

To address this problem, working electrodes need to be frequently refreshed in order to keep a clean and unpolluted surface. Toward this end, the contaminated surface of working electrodes normally should be physically removed for reproducible electrochemical detection results. This process is time-consuming and requires experienced operation so that practical applications of electrochemical detection have been largely limited. Therefore, it is of importance to develop low cost working electrodes for one-time use [3–5].

During the past years, there have been a number of studies on disposable working electrodes for a variety of purposes [6–9]. The most successful disposable working electrodes were screen-printed electrodes because of their simple fabrication and changeable formats [10,11]. Meanwhile, carbon, gold and other nanomaterials have been used for preparation of disposable electrodes with various routes [6,12–14]. For example, disposable carbon electrodes have been reported for liquid chromatographic determination

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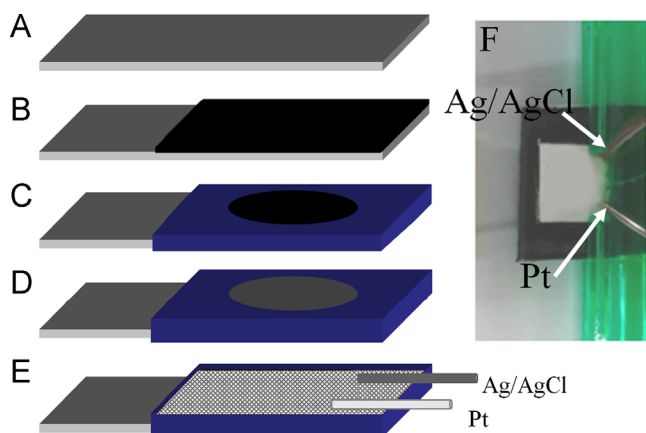


Fig. 1. The schematic of the bismuth modified carbon tape electrode and its integration with PAD for Pb detection. (A) ITO glass; (B) a piece of carbon tape was attached on the ITO glass; (C) the insulate plastic adhesive tape with a hole (diameter: 4 mm) was attached on the carbon tape; (D) the bismuth film was electrodeposited on the exposed part of the carbon tape to form the bismuth modified carbon tape electrode; (E) a piece of filter paper with 6 mm long and 6 mm wide, an Ag/AgCl wire and a Pt wire were integrated with the bismuth modified carbon tape to form the PAD for electrochemical detection and (F) the photo of the PAD.

of catecholamines in blood plasma samples based on sputtering carbon on a polyether-etherketone (PEEK) film [15]. Indium tin oxide (ITO) glass was also used as the substrate for the modification of glucose oxidase on gold nanoparticles for rapid glucose measurement [16]. It should be emphasized that the price of ITO glass has significantly decreased with the development of modern display technology. However, currently most of disposable electrodes still suffer from expensive devices, time-consuming pretreatment or unsatisfactory reproducibility. It is believed that engineered materials would be more preferable for the fabrication of disposable electrodes.

Stripping analysis has been extensively investigated for the detection of trace heavy metal ions and biochemical analysis [17,18]. Initially, mercury (such as hanging mercury electrodes) was used as the material of working electrodes since it could be refreshed conveniently for reproducible detection [19,20]. However, the strong toxicity of mercury makes its applications unfriendly to the environment. To address this problem, researchers have pursued alternative materials, such as bismuth and other materials, etc., to modify electrodes for stripping voltammetry [5,21–26]. The advantages of bismuth film include low toxicity, wide potential range, and insensitivity to dissolved oxygen. On the other hand, various materials, including graphite [27], pencil [8], carbon paste [28,29] and gold [30,31], have been used as the substrate of working electrodes in stripping analysis, which is more active than in other electrochemical analytical approaches because of its well-know mechanisms. The development of low-cost disposable working electrodes could considerably shrink the gap between fundamental research and practical applications of stripping analysis and other electrochemical detections.

In our previous report, double-sided conductive adhesive carbon tape supporting on ITO glass was used as the substrate of working electrodes in order to stabilize electrochemiluminescence emission from quantum dots [32]. In this paper, we regulated the areas of the carbon tape electrodes and then modified them with bismuth for stripping analysis. Our experimental results showed that the bismuth modified carbon tape electrodes could be used to effectively differentiate Zn, Cd and Pb ions in the paper-based analytical devices. Specifically, the disposable working electrodes could be used to quantify the concentration of Pb ions migrated from toys with similar results obtained using atomic absorption spectrometry (AAS). Our investigation implied that carbon tape

might be a potential candidate for the fabrication of low cost disposable electrodes.

2. Materials and methods

2.1. Chemicals and materials

All the chemicals were of analytical grade unless otherwise mentioned. $\text{Pb}(\text{NO}_3)_2$ and $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ were from Runjie Chemical Reagent Company (Shanghai, China). Sodium acetate anhydrous (CH_3COONa) was from Xilong Chemical Co., Ltd (Guangzhou, China). Acetic acid was from Shenbo Chemical Co., Ltd (Shanghai, China). The double distilled water was collected from SZ-93A automatic double water distiller made by Shanghai Yarong Biochemical Instruments (Shanghai, China). The stock solution of $\text{Pb}(\text{II})$ ($1000 \mu\text{g L}^{-1}$) was prepared using acetate buffer solution (0.1 M, pH 4.5) as the supporting buffer and diluted to be certain concentrations. The qualitative filter papers (Whatman No. 1) were from Whatman International Ltd. (Maidstone, United Kingdom). The Indium tin oxide (ITO) conductive glass ($355.6 \times 406.4 \times 1.1 \text{ mm STN}$, 10Ω) was purchased from Nanbo Display Technology Co. LTD (Shenzhen, China). The conductive double-sided carbon adhesive tape (12 mm wide, 0.16 mm thick and 20 m long) was purchased from SPI Supplies (West Chester, PA, USA). Toy samples consisting of Pb were obtained from Chinese Academy of Inspection and Quarantine.

2.2. Sample preparation

The toys were treated according to the national safety technical regulation for toys of China (GB 6675-2003). Briefly, they were cut or crushed at room temperature and then mixed with 0.07 M HCl at $37 \pm 2^\circ\text{C}$. Then the mixture was shaken for 1 min. After that, 2 M HCl was added to adjust the pH value to be 1.0–1.5. The mixture was stirred for 1 h and kept for 1 h at $37 \pm 2^\circ\text{C}$ without light. Then the mixture was filtrated with a membrane filter and the sample solutions were mixed with acetate buffer solution (0.1 M, pH 4.5) for detection with stripping voltammetry and AAS, respectively.

2.3. The bismuth modified carbon tape electrode

As shown in Fig. 1, ITO glass was cut to be 20 mm long and 7 mm wide. Then the conductive carbon tape (12 mm long and 7 mm wide) was attached on the conductive surface of ITO glass. A section of plastic adhesive tape was punched with a hole (4 mm diameter) and then attached on the carbon tape in order to provide an identical area for detection. The carbon tape electrode was then electrodeposited with bismuth using $60 \mu\text{l}$ of 60 mg L^{-1} bismuth nitrate in 1.5 mol L^{-1} HCl solution containing 0.15 mol L^{-1} sodium citrate by applying the potential of -1.8 V for 120 s [33]. We found that the selection of the buffer solution is critical in order to fully solve bismuth. After that, the bismuth modified carbon tape electrode was rinsed with double distilled water and dried for use.

2.4. Stripping voltammetry and other analysis

Stripping voltammetry was carried out on a CHI1230B electrochemical working station (CH Instrumentation, Shanghai, China) with a three-electrode system using the modified carbon tape electrode as the working electrode, an Ag/AgCl wire as the reference electrode and a platinum wire as the counter electrode. A piece of the Whatman filter paper No. 1 with 6 mm long and 6 mm wide was put above the surface of the bismuth modified carbon tape electrode. Sample solutions in 0.1 M acetate buffer

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