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Biomass reinforced graphene oxide solid/liquid phase membrane extraction for the measurement of Pb(II) in food samples

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ABSTRACT

Here, a solid/liquid phase extraction mode was used for the preconcentration of Pb(II) in water and food samples through a biomass reinforced graphene oxide (BGO) membrane. Taking advantage of the two modes, the synergistic effect of BGO membrane (solid extraction) and organic solvent (liquid extraction) enhances the extraction efficiency toward target ion. In detail, the effect of composition parameters in BGO membrane and experiment conditions such as pH, eluent types, elution time and sample volume were optimized, as well as shows no obvious interference toward different competing ions. Under the optimal experiment conditions, the limit of detection, precision as RSD% of this method were found to be 0.84 $\mu g\,L^{-1}$ and 4.65%, respectively. Moreover, the large enrichment factor of BGO membrane demonstrates the applicability of the use of large sample volumes. Furthermore, the method was successfully verified by analyzing spiked Pb(II) in water and food samples.

1. Introduction

Lead pollution in the environment and food chain system is known to be dangerous for flora, fauna as well as human health. The non-biodegradable Pb(II) is explicitly toxic in small amounts, which can cause poisoning symptoms for human health including abdominal pain, headaches, convulsions, anemia, chronic nephritis, brain damage, and central nervous system disorders (Baranik et al., 2018; Wang, Zhang, Wang et al., 2016; Yang, Wang, Chen et al., 2018). According to US EPA regulations for safe drinking water, the maximum concentrations of 15 $ng\,mL^{-1}$ cannot be exceeded (United States Environmental Protection Agency: Washington). In food matrix, the stringent restrictions are also exist such as the maximum tolerable concentration limit in fruit juice is 0.05 $mg\,kg^{-1}$ (Codex Alimentarius, 1995). Therefore, the determination of this highly toxic metal in environmental and food samples is a matter of great importance. Nevertheless, the direct determination of ultratrace amounts of Pb(II) in complex matrixes is difficult thus the use of preconcentration methods is necessary.

Among various techniques, solid phase extraction (SPE) and liquid phase extraction (LPE) are widely accepted for their unparalleled advantages of low consumption of organic solvent, high enrichment efficiency and simple operation, which simplifying the sample pretreatment and have been widely used for the analysis of environmental, clinical, food, forensic, biological, and pharmaceutical samples with or without derivation (Liu et al., 2017; Li et al., 2018; Samsidar, Siddiquee, & Shaarani, 2017; Salazar-Beltrán et al., 2018). However, each of them has its own disadvantages: the main drawback of SPE is related to the limited reproducibility and nonsufficient enrichment; LPE may suffer from some drawbacks such as the need of large amount of toxic organic solvent and the requirement of time consuming, tedious and multistage operation (Bahar, Es'haghi, Nezhadali, Banaei, & Bohlooli, 2017). To date, there have been reported a so-called supported solid/liquid phase microextraction method, which was proposed by filling small amount of an organic phase and nanoadsorbent into the lumen and pores of hollow fiber that act as a support for two-phase operation (Bahar et al., 2017). The synergistic effect of solid (adsorbent as analyte trapper) and liquid phase (extract solvent) increases the amount of analyte extracted and improves the extraction efficiency by reducing the Nernst diffusion layer, which makes it an attractive method for the microextraction of diverse analytes (such as toxins, pharmaceuticals and heavy metal ions) with qualified high pre-concentration factor (Es'haghi et al., 2010; Song et al., 2012; Sehati et al., 2014; Wang et al., 2017). However, it still suffers from the limited applicability for only small sample volumes, which is difficult to

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achieve efficient extraction in high sample volumes with low concentration of target ions in practical utilization.

Generally, solid phase extraction is performed by passing sample solution through supported sorbent in the format of column, cartridge or membrane disk (Wang et al., 2018). Of these, SPE membrane disk with high surface area and uniform particle distribution can make extraction efficiency comparable with other forms (Zhou et al., 2017). Moreover, it provides much greater cross-sectional areas than the cartridge or column, which make the utilization of high flow rate and large sample volumes possible (Leandro et al., 2006). Considering the effectiveness of simultaneous two-phase extraction in enhancing the target extraction, we hypothesize the development of novel membrane architecture assisted of organic solvent treatment is of great importance to allow efficient extraction for practical operation in two-phase modes. Since this extraction mode allows the simultaneous extraction of the target ions from the sample solution to solid/liquid media and the subsequent transference of the analytes to an aqueous acceptor phase.

Based on the above statement, here, we developed a biomass reinforced graphene oxide (BGO) membrane for solid/liquid phase extraction (SLPE) of Pb(II) in water and food samples. In this BGO-SLPE method, two extractants including organic solvent (liquid extraction medium) and all carbon-based membrane (solid extraction one), in which the later was considered as support for the former, are worked together to implement an effective extracting strategy. In detail, to evaluate the analytical performance of the membrane-based SLPE method, Pb(II) was used as the target analyte. The composition parameters in BGO membrane influencing the recoveries of the targets were optimized in detail. The effects of various parameters such as pH, eluent types and elution time, sample volume, and interfering ions on the recoveries of the analytes were also investigated. Moreover, the as-prepared BGO membrane was also evaluated for the extraction of Pb^{2+} in water and food samples.

2. Experimental

2.1. Reagents and materials

The peanut shells were collected from the commercial supermarket. All reagents were of analytical grade. The reagents such as potassium oxalate (≥99.8%) and calcium carbonate (≥99.0%) were supplied by Sinopharm Chemical Reagent Co.. Different initial concentrations of Pb^{2+} solution were prepared by dissolving $Pb(NO_3)_2$ (Sinopharm, 99.99%) in deionized water. All experiments and solution preparations were carried out using deionized water.

2.2. Physical instrumentation

Scanning electron microscopy (SEM) images were recorded on a Hitachi S-4800. The concentration of anions extract from the monolith column was determined by flame atomic absorption spectrometer (FAAS) (ZEEnit700P, Analytikjena). The wavelength selected for the determination of the analyte was 283.3 nm for Pb, using an air-acetylene flame burner.

2.3. Preparation of BGO membrane

In this study, GO was synthesized from graphite powder *via* a modified Hummers method as described in reported work (Yang, Wang, Zhang et al., 2017). The CKP biomass was prepared from the peanut shell and fabricated by Sevilla et al. work as: the carbon precursor of the peanut shells was mixed with potassium oxalate and calcium carbonate (weight ratio = 1:1:1) and subjected to heat-treated at 800 °C (5 °C min^{-1}) under a nitrogen gas flow for 1 h. Then the prepared products were washed by diluted HCl for three times and dried overnight at 60 °C (Sevilla et al., 2017).

To fabricate the all-carbon nanoarchitectured BGO membranes, 50 mL diluted GO solution (0.1 mg mL^{-1}) was first prepared from the stock and subjected to ultrasonication for 30 min to obtain the exfoliated GO sheets. Then, 20 mg of CKP biomass was dispersed in GO solution *via* ultrasonication for 1 h to give well-dispersed mixture solutions. Finally, the obtained mixtures were subjected to pressure-assisted filtration at 1 bar pressure on polycarbonate (PC) membranes (0.22 μm pore size, 47 mm diameter, Millipore) using a dead-end filtration device.

2.4. Solid/liquid phase extraction method with BGO membrane

Before each sample loading, all of the BGO membranes were immersed in the extract solvent of caprylic acid for 30 min. The pretreated sample solution was react with the pretreated BGO membrane for 10 min then passed through it by vacuum-assisted filtration. After that, the membranes were eluted with 2.0 mL eluent solvent for FAAS analysis. Each extraction was carried out in three replicates for parallel test.

2.5. Experimental design and data analysis

All processes were carried out in three replicates. The removal efficiency of BGO membrane toward Pb^{2+} was calculated according to the following equation:

$$Removal(\%) = (Co - Ce)/Co \times 100\% \quad (1)$$

where C_0 and C_e represent the initial and permeate concentrations of Pb^{2+} aqueous solution (g L^{-1}), respectively.

2.6. Preparation of water and food samples

To evaluate the BGO-SLPE method, the samples of orange juice, water (tap water, and mineral water) and liqueur were analyzed. Drinking samples (2 mL) were placed into 20 mL beakers, then 3 mL concentrated HNO_3 (65%, w/w) and 3 mL $HClO_4$ were added, respectively. The mixture was digested under 80 °C for 20 min, and then the solution was heated on a hot plate at 200 °C until a clear solution was obtained. Finally, it was diluted with deionized water to 50 mL. The digestion of pork liver sample (0.5 g) was treated as same as the drinking samples. The tap water and mineral water were used without the digestion procedure.

3. Results and discussion

3.1. Characterization of BGO membrane

BGO membranes were successfully assembled through vacuum-assisted filtration of the mixture of biomass and GO onto the support substrate of 0.22 μm PC membrane, as schematic illustrated in Fig. 1a. The fabrication process began with the preparation of CKP using a one-pot pyrolysis method, which was calcinated at 800 °C in the presence of potassium oxalate (activating agent) and calcium carbonate (hard template) as reported before (Sevilla et al., 2017). CKP biomasses were then intercalated between the GO sheets (Fig. 1b) to create nanochannels in order to mitigate the drop in water permeability brought by the decreased in the interlayer spacing due to the π-π interaction between interlayer GO nanosheets, which facilitate water transport through the ultrathin GO membranes. The magnified surface and cross-section morphology of pure GO and BGO membrane were observed using SEM. The pure GO membrane shows completely smooth surface and densely interlayer distance (Fig. S1), while the BGO membrane displays highly wrinkled surface and enlarged interlayer distances with biomass particles intercalation as shown in Fig. 1d and e. Moreover, the XRD patterns in Fig. S2 shows the pristine GO revealed an intense, sharp peak centered at 2θ = 11.31°, corresponding to (0 0 2) peak of GO sheets and the interplanar spacing is 0.88 nm (Xu, Wang, Zu, Han, & Wei, 2010). In the case of BGO membrane, the characteristic Bragg

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