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Graphene oxide-based dispersive micro-solid phase extraction for separation and preconcentration of nicotine from biological and environmental water samples followed by gas chromatography-flame ionization detection

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ABSTRACT

Graphene oxide (GO) has showed great potential to use as an adsorbent in sample preparation procedures. In this research, GO was used as an effective adsorbent in a simple GO-based dispersive micro-solid phase extraction (GO-D-μ-SPE) method for isolation and preconcentration of nicotine prior to gas chromatography-flame ionization detection (GC-FID). The prepared GO was characterized by X-ray diffraction (XRD), Fourier transform infrared (FT-IR) spectroscopy, scanning electron microscope (SEM), thermogravimetric analysis/differential thermal analysis (TGA/DTA), and ultraviolet–visible (UV–vis) absorption spectroscopy techniques. Various experimental parameters affecting the extraction recovery, including the amount of GO, extraction time, pH of the sample solution, salt concentration, and desorption conditions were investigated and optimized. Under the optimized conditions, a linear response was obtained in the concentration range of 5–2000 ng mL^{-1} with a determination coefficient of 0.9987. The limit of detection (LOD) of the method at a signal to noise ratio of 3 was 1.5 ng mL^{-1}. The linearity was in the concentration range of 5–2000 ng mL^{-1} with a determination coefficient of 0.9987. Intraday and inter-day precisions were obtained equal to 2.7% and 5.2%, respectively. The method was successfully applied to the nicotine analysis in biological and water samples with the recoveries in the range of 88.7–109.7%.

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1. Introduction

Nicotine, 3-(1-methyl-2-pyrrolidinyl) pyridine, is a highly toxic alkaloid with a lethal amount of 30–60 mg kg^{-1}. It is found naturally in high levels (2–8%) in tobacco leaves. Nicotine is the primary substance that causing addiction to smoking. It can be absorbed immediately in humans through the skin, mucosal lining of the mouth and nose or by inhalation in the lungs, which results in an increase in blood pressure and heart rate [1]. Furthermore, it is suspected to contribute to some damages such as cardiovascular disease, kidney disease and cancer [2,3].

Owing to high solubility in water, transfer of nicotine to surface waters through the wastewater system of different industries such as tobacco processing, cigarette manufacturing [4,5] and pharmaceutical industry [6–8] causes considerable environmental pollution. Besides, it has been used for a long time as a pesticide in agriculture due to its easy and low cost extraction from available natural resources such as tobacco leaves. In the European Union, all applications consisting of nicotine as a pesticide have been banned since 2009. However, it is still used in many developing countries to control pests [9–11]. Thus, the development of sensitive and specific analytical methods for the extraction and detection of trace amounts of nicotine in biological and environmental samples has become one of the major interest.

Trace analysis of analytes in various types of samples generally requires a pretreatment step to separate and enrich them before instrumental determination. Several sample pretreatment methods such as liquid–liquid extraction (LLE) [12–14], cloud point extraction (CPE) [15], pressurized liquid extraction (PLE) [16,17], liquid-phase microextraction (LPME) [18], solid phase extraction (SPE) [19–21], solid-phase microextraction (SPME) [22–24] and single drop microextraction (SDME) [25] have been reported for isolation and preconcentration of nicotine from different matrices prior to gas chromatography (GC) or liquid chromatography (LC) analyses. Among these methods, SPE is widely used due to its simplicity, reproducibility and availability. In a SPE procedure, the sorption of analyte(s) on an appropriate solid sorbent and the subsequent desorption using a suitable solvent, eliminate the potential interferences and preconcentrate analyte(s) of interest.

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The sorbent materials used in SPE should provide the requirements for a good sample pretreatment (e.g. appropriate attraction, high sorption capacity, good recovery and enrichment factor) [26]. Therefore, great efforts are continuously being made to develop new materials for this purpose.

Nowadays, nanoscale carbon-based materials, such as fullerene, carbon nanotubes, graphene and graphene oxide have attracted considerable attention in SPE for the isolation or extraction of various compounds, due to their large surface area, chemical and thermal stability, ease of surface functionalization or modification and excellent adsorption capacity [27–32]. Graphene oxide (GO) is the oxidized derivative of graphene which is usually produced through the strong oxidation of graphite [33]. The GO sheets consist of a hexagonal ring-based carbon network that are covalently bonded with oxygen functional groups (such as hydroxyl, epoxy, and carboxyl). The oxygenated lattice of GO provides good solubility and dispersibility of this material in many solvents, particularly in water and non-covalent interaction with various compounds through electrostatic interaction and hydrogen bonding. Furthermore, the two dimensional plane structure and single-atom thickness of GO possesses an ultra-high specific surface area and high adsorption capacity [29,34]. These considerable properties, give GO great ability to use as a sorbent material in extraction/preconcentration techniques.

In this work, a simple method for the analysis of nicotine in biological and environmental water samples was established based on the GO-dispersive micro-solid phase extraction (GO-D-μ-SPE) in which a dispersive system based on rapid injection of GO aqueous solution into the sample solution was applied. To the best of our knowledge, this may be the first report about the use of graphene oxide as a novel sorbent for the isolation and preconcentration of nicotine. The effects of the adsorption and desorption factors on the extraction recovery of the analyte were studied systematically.

2. Experimental

2.1. Reagents and materials

Sulfuric acid (99.8%), hydrogen peroxide (H_2O_2, 30%), hydrochloric acid (HCl, 37%), quinoline, triethylamine (TEA), acetonitrile (ACN), ethyl acetate (EA), dichloromethane (CH_2Cl_2), sodium chloride (NaCl), sodium hydroxide and potassium permanganate ($KMnO_4$) with the purity higher than 99.9% were obtained from Merck Chemicals (Darmstadt, Germany). Methanol (MeOH), was obtained from Sigma Aldrich Ltd (St Louis, USA). Graphite powder was purchased from Samchun Pure Chemical Co Ltd (Pyeongtaek, Korea). Nicotine ($\geq$ 99%) was purchased from Acros Organics (New Jersey, USA). Nicotine and quinoline (internal standard) stock solutions were prepared in methanol at the concentration level of 10 mg mL^{-1} and stored at 4 °C before use. The working solutions were daily prepared by subsequent dilution of stock solution in deionized water.

2.2. Instrumentation

Gas chromatography. The GC analyses were carried out on a Shimadzu GC-17A (Tokyo, Japan) gas chromatograph equipped with a FID detection system and a split/splitless injector. The separations were performed with a CBP-5 capillary column (25 m, length; 0.25 mm internal diameter; 0.22 μm, film thickness; stationary phase, 5% phenyl–95% methyl polysiloxane). The carrier gas was helium with the purity of 99.999%. The injection port was set at 240 °C and used in the splitless mode at the splitless time of 0.30 min with the split ratio of 1:50. A Shimadzu OPGU-2200s hydrogen generator (Tokyo, Japan) was used to supply H_2 (g) for FID. The detector temperature was adjusted at 260 °C. The oven temperature was programmed as follows: initial temperature 100 °C (held for 1 min) raised to 260 °C at a rate of 30 °C min^{-1} (held for 5 min).

The pH values were measured using a WTW Inolab 720 pH meter (Weilheim, Germany). The centrifuges were performed with a Hermle centrifuge model Z 200 A (Wehingen, Germany). An Eurosonic 4D (Euronda, Montecchio Precalcino (Vincenza) Italy) ultrasonic water bath and a vortex mixer model ZX-Classic (Velp Scientifica, Milan, Italy) were employed for homogenization of solutions.

The infrared transmittance spectra were recorded using an Equinox 55 FT-IR spectrometer (Bruker, Bremen, Germany) in the range of 400–4000 cm^{-1}. The X-ray diffraction (XRD) patterns were collected on a X'Pert Pro MPD X-ray diffractometer (Almelo, Netherlands) with a Cu K_α radiation (λ = 1.54178 Å). The morphology of GO was observed by scanning electron microscopy (SEM) with a Hitachi S-4160 machine (Tokyo, Japan) at an accelerating voltage of 20 KV.

2.3. Synthesis of graphene oxide

Graphene oxide was synthesized based on Hummers' method [35]. Firstly, 2.0 g graphite powder was added to 100 mL concentrated H_2SO_4 containing 1.5 g of $NaNO_3$ in a 500 mL flask. The mixture was allowed to react in an ice bath with constant stirring for 2 h. A portion of $KMnO_4$ (12 g) was then gradually added to the mixture under vigorous stirring. During this step, the temperature was carefully controlled to not exceeding 10 °C. The resulting suspension was stirred at 40 °C for 24 h, and then diluted slowly by 100 mL of deionized water (the temperature did not exceed 40 °C). Subsequently, it was heated to 95 °C and kept in this temperature for 15 min. Afterward, 300 mL deionized water was added to the mixture to stop the reaction. Successively, 20 mL of H_2O_2 was added dropwise to the mixture to reduce the residual $KMnO_4$. The resulting product was washed several times with 5% HCl aqueous solution (1000 mL) to remove impurities and sulfate ions, and then with distilled water to remove excess acid. Thereafter, the rinsed product was dispersed in deionized water using an ultrasonic bath for 30 min and finally air-dried under ambient conditions. The obtained GO was characterized and used in the following experiments.

2.4. Real sample collection and preparation

Urine and saliva samples were kindly collected from smoker and non-smoker volunteers in our laboratory and stored in polypropylene tubes at −20 °C. Prior to the sample preparation step, the samples were defrosted and centrifuged at 5000 rpm for 5 min to remove any particulate matter. Then, proteins were precipitated by addition of 1 mL ACN to 1 mL of sample solutions with subsequent vortexing for 5 min. The mixtures were maintained for 20 min at 4 °C and then centrifuged at 5000 rpm for 5 min. The upper phase was then separated and diluted to 10 mL with distilled water.

Water samples were collected from the Darband and Darakeh Rivers in north of Tehran (capital of Iran). There were many hookah lounges alongside the rivers' banks where their wastes were directly introduced to the rivers and cause a huge amount of pollutants including nicotine. The collected water samples were filtered through a 0.45 μm membrane filter and stored in brown glass bottles at 4 °C until used for analysis.

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