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Review

Graphene and carbon nanotubes as solid phase extraction sorbents for the speciation of chromium: A review



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HIGHLIGHTS

GRAPHICAL ABSTRACT

- Carbon nanomaterials (graphene and CNTs) as sorbents in solid phase extraction.
- Solid phase extraction strategies for Cr speciation with graphene and CNT as sorbents.
- Last trends for Cr speciation with functionalized graphene and CNTs.

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ABSTRACT

In the last decades, the extensive use of chromium in industrial activities has led to the discharge of different chromium species into the biosphere. The two stable chromium forms are Cr(III) and Cr(VI), which have dramatically different properties. While the first one is essential, the second is harmful and carcinogenic, even at very low concentration. Therefore, the appropriate analysis of chromium in environmental, biological, food and other kind of samples need a reliable separation and subsequent quantification of both Cr species. The present paper provides a critical review of chromium speciation methods in which solid phase extraction was employed as sample pretreatment using graphene and carbon nanotubes (and their diverse oxidized, functionalized and magnetic derivatives) as sorbents. The different published methods for the speciation of Cr(III), Cr(VI) or both species are described and

ABBREVIATION: 2-5-Br-PADAP, 2-(5-bromo-2-pyridylazo)-5-diethylaminophenol; AAPTS, 3-(2-aminoethylamino)propyltrimethoxysilane; APDC, ammonium pyrrolidine dithiocarbamate; CNF, carbon nanofiber; CNT, carbon nanotube; CRM, certified reference material; CTAB, cetyl trimethyl ammonium bromide; D2EHPA, di-(2-ethylhexyl) phosphoric acid; DCC, N,N'-dicyclohexylcarbodiimide; DDTC, diethyldithiocarbamate; DPC, diphenylcarbazone; DSPE, dispersive solid phase extraction; EDA-MWCNT, ethylenediamine-modified multi-walled carbon nanotube; EDXRF, energy dispersive X-ray fluorescence spectrometry; ETAAS, electrothermal atomic absorption spectrometry; FAAS, flame atomic absorption spectrometry; FIA, flow injection analysis; G, graphene; GO, graphene oxide; GO-MWCNT-DETA, diethylenetriamine-functionalized carbon nanotubes dispersed in graphene oxide colloids; ICP, inductively coupled plasma; ICP-MS, inductively coupled plasma mass spectrometry; ICP-OES, inductively coupled plasma optical emission spectrometry; IDA, iminodiacetic acid; IUPAC, International Union of Pure and Applied Chemistry; LIF, laser induced fluorescence; LOD, limit of detection; m-GO/SiO2@coPPy-Th, SiO2-coated magnetic graphene oxide modified with polypyrrole-polythiophene; m-GO, magnetic graphene oxide; m-GO@PANI, polyaniline-coated magnetic graphene oxide; m-GO-TETA, graphene oxide decorated with triethylenetetramine-modified magnetite; MIP, molecular imprinted polymer; m-MWCNT, magnetic multi-walled carbon nanotube; MnO₂/Fe₃O₄/o-MWCNTs, manganese dioxide/iron oxide/acid oxidized multi-walled carbon nanotube magnetic nanocomposite; m-SPE, magnetic solid phase extraction; MWCNT, multi-walled carbon nanotube; MWCNT-AAPTS, 3-(2-aminoethylamino)propyltrimethoxysilane-modified MWCNT; MWCNT-AP-Modified-SiO₂, aminopropyl-modified silica carbon nanotube; NIOSH, National Institute for Occupational Safety and Health; PAHs, polycyclic aromatic hydrocarbons; PANI, polyaniline polymer; PDDA, poly(diallyldimethylammonium chloride); P-EDXRF, polarized energy dispersive X-ray fluorescence; PF, preconcentration factor; S-CS-MWCNTs, Schiff base-chitosan-grafted multi-walled carbon nanotubes; SDS, sodium dodecyl sulfate; SIA-BI-SPE, sequential injection analysis-bead injectionsolid phase extraction; SPE, solid phase extraction; SWCNT, single-walled carbon nanotube; TEOS, tetraethyl orthosilicate; TETA, triethylenetetramine; TRG-SiO₂-APTES, thermally reduced graphene modified silica-supported 3-aminopropyltriethoxysilane; TRG, thermally reduced graphene; TRXRF, total reflection X-ray fluorescence spectrometry; XRF, X-ray fluorescence spectrometry.

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Solid phase extraction Chromium speciation classified on the basis of the separation strategy employed: (i) the selective retention of only one of the two species by the sorbent, (ii) the retention of a Cr-chelate formed by using a ligand selective for only one of the Cr-species, or (iii) the sorption of both Cr(III) and Cr(VI) species. In addition, the distinct applied SPE modes (on-column, dispersive or magnetic) and their automation possibilities, the analytical techniques utilized for measurement of the Cr-species after separation, as well as the analytical figures of merit of the methods developed up to date are evaluated. Finally, the expected future trends of solid phase extraction for Cr speciation based on carbon nanomaterials such as graphene, carbon nanotubes and their derivatives as sorbents are commented.

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Contents

1.	1. Introduction		2
2. Carbon nanotubes and graphene as SPE sorbents		on nanotubes and graphene as SPE sorbents	3
3.	Chromium speciation using CNT- and graphene-based SPE		3
	3.1.	Chromium speciation strategies	4
	3.2.	Chromium speciation methods	6
		3.2.1. Methods for speciation analysis of Cr(III) only	6
		3.2.2. Methods for speciation analysis of Cr(VI) only	8
		3.2.3. Methods for the speciation of the element	11
	3.3.	SPE modes and automation	12
	3.4.	Analytical techniques and figures of merit	13
4.	Concluding remaks and future trends		. 14
	Refer	ences	15

1. Introduction

Natural sources of chromium include weathering processes on rocks that are rich in this element, emanations from volcanos, and soil erosion. However, the highest proportion of this element in the environment is generated from anthropogenic sources. Chromium is a heavy metal that is widely used for several applications in the metallurgy industry, in the chemical production of paints and pigments, in synthetic fertilizers and crop protection products, and in leather tanning activities, amongst others [1]. As a consequence of these human activities different chromium species are discharged into the environment. In general, Cr can exist in different chemical oxidation states from 0 to VI, but in the environment the only stable forms are Cr(III) and Cr(VI). Both species have dramatically different properties. Cr(III) is an essential form of this element that play important roles in the metabolism of glucose, lipids and proteins in living organisms. In fact, the recommended human intake for Cr(III) is 50–200 μ g day^{-1.} In contrast, Cr(VI) is a highly toxic species and it produces adverse carcinogenic and mutagenic effects in cells [2–4]. The general population may be in contact with this species mainly through inhalation of Cr(VI) compounds in polluted air and by ingestion of contaminated water. In fact, in different countries diverse regulations have been adopted that limit the content of total chromium (Cr_{TOT}) and Cr(VI) in water and in air. In the case of Europe, EC Directive 98/83/EC [5] established maximum amounts of Cr_{TOT} and Cr(VI) in drinking water of 50 and 20 µg L⁻¹, respectively. The US Environmental Protection Agency limited the maximum contaminant level for Cr_{TOT} in water to 100 μ g L⁻¹, but the Office for Environmental Health Hazards Assessment of California established, in July 2011, a public goal of 0.02 μ g L⁻¹ for Cr(VI) as a protection level against the undesirable toxic effects of this pollutant [6]. Furthermore, in working ambient air NIOSH recommends that airborne exposure to all Cr(VI) compounds be limited to a concentration of 0.2 μ g Cr(VI) m⁻³ for an 8-h time-weighted average exposure during a working week of 40 h [7].

On the basis of the above considerations, it is clear that the development of appropriate methods for the determination of chromium species is an analytical challenge. The separation of Cr(III) and Cr(VI) prior to analytical measurement in speciation studies has been carried out using a range of sample pretreatment procedures (ion exchange, precipitation, liquid-liquid extraction, etc.) [8]. However, solid phase extraction (SPE) has proven to be one of the most attractive alternatives for this goal with the double objective of separating and concentrating the chromium species and removing the sample matrix (as well as possible interferences). The advantages of SPE include the high preconcentration factor, good recovery, rapidity, use of small quantities of organic solvents, variety of sorbents and the possibility of automation of the whole pretreatment process [9,10]. Therefore, SPE has been employed for Cr speciation studies using different types of sorbents (such as diverse type of functionalized resins and ion-exchangers; chitosan, alumina, silica and derivatives; inorganic solids; C8 and C18 bonded-silica, etc.). Interesting critical reviews and research papers have been published describing pretreatment procedures (including sorbent types employed), species, sample type, techniques and figures of merit of the proposed methodologies for Cr speciation in different solid and liquid matrices [11–14]. Nowadays, carbon-based nanomaterials, mainly graphene and carbon nanotubes, have proven to be useful sorbents in SPE because of their physical and chemical properties. From this point of view, these two materials have been proposed and used as SPE sorbents for Cr speciation in the last few years. Taking into account that graphene and carbon nanotubes (and derivatives) can be promising sorbents for such task, in this work, a survey on the concrete methodologies using these carbon nanomaterials as SPE sorbents for the speciation of chromium is presented. Different speciation strategies, diverse functionalizations, different SPE modes employed, analytical characteristics of the published methods and future trends in this field are discussed in this review.

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