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Nanoparticle-assisted chemical speciation of trace elements

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ABSTRACT

Speciation analysis of trace elements is a relevant topic with involvements in many areas, e.g. environment, food, biology, medicine, etc. Nanomaterials have mainly found application in speciation analysis taking advantage of their sorptive, catalytic or optical properties. Different kinds of nanosorbents (metal oxides, carbon nanotubes, fullerenes, noble metals, etc.) have contributed to achieving simple and efficient speciation strategies in combination with atomic spectrometry. The design of new interfaces involving photocatalysts such as nano-TiO_2 has enabled a remarkable simplification of hyphenated techniques for speciation by promoting both photooxidation and photoreduction reactions. Finally, plasmonic (e.g. AuNPs and AgNPs) and fluorescent nanoparticles (e.g. quantum dots, carbon dots) have provided selective and sensitive assays for optical sensing of different elemental chemical forms.

In this overview, we offer an insight into the analytical potential of different nanomaterials for chemical speciation of trace elements.

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Abbreviations: AAS, atomic absorption spectrometry; AFS, atomic fluorescence spectrometry; APDC, ammonium pyrrolidinedithiocarbamate; CD, carbon dot; CDTA, 1,2 cyclohexanediamine- tetraacetic acid; CE, capillary electrophoresis; CL, chemiluminescence; CNT, carbon nanotube; CPE, cloud point extraction; CV, cold vapour; CVG, chemical vapor generation; DDTC, diethyldithiocarbamate; DMA, dimethylarsinate; DMSPE, dispersive microsolid-phase extraction; EDTA, ethylendiaminetetraacetic acid; EF, enrichment factor; ETAAS, electrothermal-atomic absorption spectrometry; EtHg, ethylmercury; ETV, electrothermal vaporization; FAAS, flame-atomic absorption spectrometry; FI, flow injection; FRET, fluorescence resonance-energy transfer; G, graphene; GC-MS, gas chromatography-mass spcctrometry; GO, grapheme oxide; HG, hydride generation; HPLC, high performance liquid chromatography; IC, ion chromatography; ICP-AES, inductively coupled plasma-atomic emission spectrometry; ICPMS, inductively coupled plasma mass spectrometry; IEP, isoelectric point; LDH, layered double hydroxide; LC, liquid chromatography; LLLME, liquid-liquid-liquid microextraction; LOD, limit of detection; LOV, lab-on-valve; LSPR, localized surface plasmon resonance; MeHg, methylmercury; MEKC, micelle electrokinetic capillary chromatography; MMA, monomethylarsonate; MMT, methylcyclopentadienyl-manganese tricarbonyl; MNPs, Magnetic nanoparticles; MPY, 4-mercaptopyridine; MESNa, 2-mercaptoethane-sulphonate; MSP, magnetic solid phase; MW, microwaves; MWCNTs, multi-walled carbon nanotubes; NaDDC, sodium diethyldithiocarbamate; NP, nanoparticle; PAN, pyridylazonafthol; PhHg, phenylmercury; PPHF, polypropylene hollow fiber; QD, quantum dot; RSD, relative standard deviation; SDME, single-drop microextraction; $SeCys_2$, selenocystine; SeMet, selenomethionine; SERS, surface enhanced raman spectroscopy; SFI, stopped flow injection; SI, sequential injection; SPE, solid-phase extraction; SWCNTs, single-walled carbon nanotubes; TBT, tributyltin; TEL, tetraethylead; TXRF, total reflection X-ray fluorescence; UV, ultraviolet; VG, vapour generation.

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1. Introduction

The topic of speciation analysis has received increased attention during the last years, since it is the basis for studies of bioavalability, mobility and toxicity of elemental species in a broad variety of research fields such as environment, occupational health, toxicology, pharmacy, medicine, nutrition, biology, etc. [1]. Given the large variety of metal species with different properties and behaviour, ranging from simple chemical forms such as oxidation states up to the most complex metal ion-binding biomolecules, a plethora of analytical strategies have arisen to cope with chemical speciation of trace elements. In line with current trends of analytical chemistry, development of simplified methods being green, cost-effective, time-saving and with the feasibility for on-site analysis is a goal to be pursued. Among the different approaches available to perform chemical speciation analysis, those avoiding tedious sample pretreatment, chromatographic steps, complex clean-up and use of complex instrumentation requiring high investment and operating costs as well as high skill level, are particularly desirable. In this sense, non-chromatogaphic speciation methods have been developed in the past for this purpose [2], mainly when samples containing a few simple chemical forms are to be analysed. In the last few years, the extension of the nano-world to analytical chemistry has yielded a number of enhancements focused on three relevant stages of the analytical process, i.e. sample pretreatment, separation and detection [3]. Different nanoanalytical approaches for chemical speciation of trace elements have been reported, with enhanced performance and outstanding simplicity as compared to earlier speciation methods. So far, most of them have focused on samples containing a reduced number of species. These include oxidation states, e.g. Cr(III)/Cr(VI), As(III)/As(V), etc., and in some cases, metal complexes such as small organo-(metal)loids, e.g. methylmercury (MeHg), tetraethyllead (TEL), selenomethionine (SeMet), monomethylarsonate (MMA), tributyltin (TBT), etc.

Several review articles have appeared in the literature dealing with applications of specific nanoparticles (NPs) for solid-phase extraction (SPE) using, e.g. carbon nanotubes (CNTs) [4], fullerenes [5], magnetic solid phases (MSPs) [6,7], TiO_2 [8], etc., and for optical sensing using plasmonic, e.g. AuNPs and AgNPs [9,10] or fluorescent nanoparticles [11,12]. Nevertheless, those reviews mostly tackled the determination of total metal contents but chemical speciation was not addressed. Recently, applications of nanomaterials for separation/preconcentration of metal species including their integration in microfluidic systems have also been reviewed [13].

As shown in Fig. 1, nanoparticle-assisted chemical speciation relies on three different kinds of nanomaterials, namely nanosorbents, nanoprobes and nanocatalysts.

Nanomaterials have been involved in several ways in order to facilitate chemical speciation, namely: i) selective sorption of chemical forms using SPE, e.g. use of Fe_3O_4, Al_2O_3, CNTs, etc. as nanosized sorbents; ii) optical sensing of metal species, e.g. use of AuNPs as colorimetric nanoprobes; iii) conversion of a chemical form to an amenable species to the anytical technique, e.g. prereduction of Se(VI) to Se(IV) and further hydride generation-atomic absorption

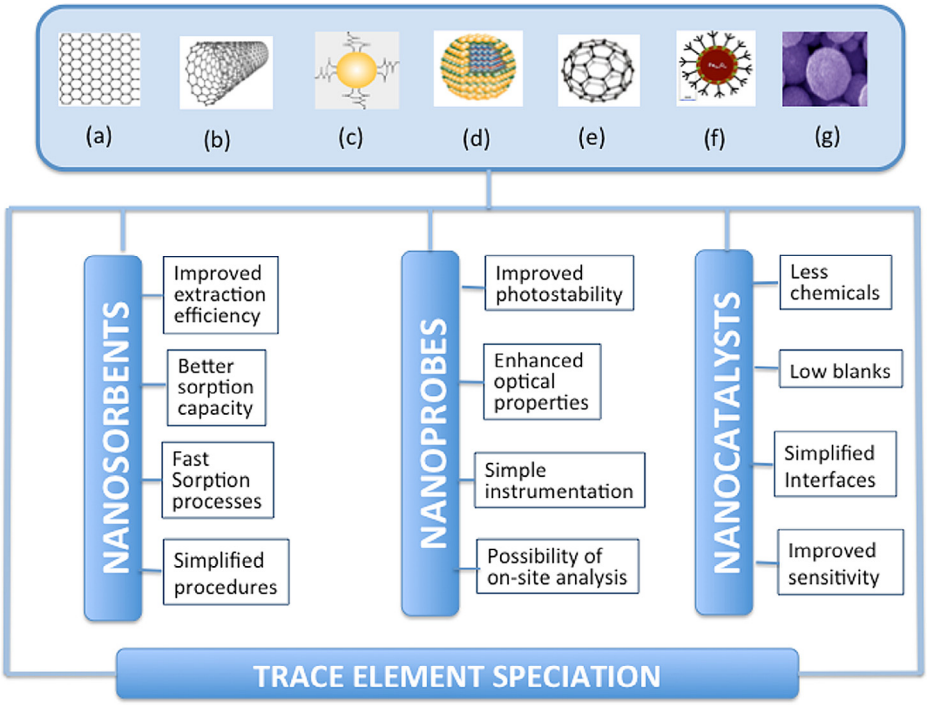

Fig. 1. Representative examples of nanomaterials involved in speciation analysis of trace elements along with main advantages provided. (a) Graphene; (b) carbon nanotube; (c) Au NP; (d) quantum dot; (e) fullerene; (f) nano-Fe_3O_4; (g) nano-TiO_2.

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