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## **CRITICAL REVIEW**

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Recent advances in the modification of electrodes for trace metal analysis: a review †

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This review paper summarizes the research published in the last five years on using different compounds and/or materials as modifiers for electrodes employed in trace heavy metal analysis. The main groups of modifiers are identified, and their single or combined application on the surface of the electrodes is discussed. Nanomaterials, film-forming substances, and polymers are among the most used compounds employed mainly in the modification of glassy carbon, screen-printed, and carbon paste electrodes. Composites composed of several compounds and/or materials have also found growing interest in the development of modified electrodes. Environmentally friendly substances and natural products (mainly biopolymers and plant extracts) have continued to be included in the modification of electrodes for trace heavy metal analysis. The main analytical performance parameters of the modified electrodes as well as possible interferences affecting the determination of the target analytes, are discussed. Finally, a critical evaluation of the main findings from these studies and an outlook discussing possible improvements in this area of research are presented.

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#### 1. Introduction

This work presents the recent developments in the research published in the last five years (i.e., 2018-2022, including the research published so far in 2023) on the modification of different types of electrodes used in trace heavy metal determination. Literature research on this area showed a large number of publications published in the last five years. As shown in

at



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#### **Critical Review**

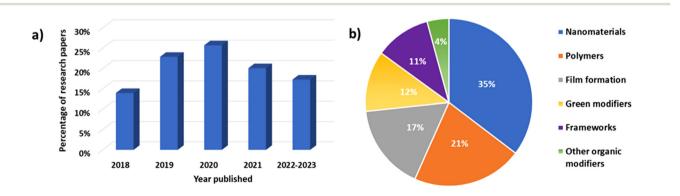
Fig. 1a, there has been continuously increased interest in the research community throughout these years (*i.e.*, an increased number of publications in 2018–2020) on the use of different types of modifiers. The decline in the research work published in 2021 and 2022 can be attributed to the slowdown from the pandemic. The focus of this review paper is to discuss the use of nanomaterials, film-forming substances (to obtain metallic or polymeric film-modified electrodes), polymers, metal- and covalent–organic frameworks, as well as environmentally friendly substances and natural products for the modification of different types of electrodes employed in trace heavy metal determination. To the best of the authors' knowledge, no comprehensive review of the research work published in this area in the last five years has been presented.

Heavy metals are naturally occurring metals and metalloids with a relatively high atomic number (higher than 20) and density (higher than 5 g cm<sup>-3</sup>).<sup>1,2</sup> Some of these elements (*i.e.*, Cu, Fe, Zn, and Fe) in small quantities are considered essential and can be part of enzymatic and metabolic mechanisms in different systems. Other metals, such as Cd, Hg, Pb, and As are not essential and show high toxicity even at lower concentrations (*i.e.*, at ppb levels).<sup>2</sup> The toxicity of these heavy metals combined with their ability to bioaccumulate and the fact that they are nondegradable present a significant threat not only to the environment (*i.e.*, water, soil, and air), but also to human health.<sup>1–4</sup>

Determination of trace level concentrations of heavy metals present in different matrixes (mainly in water) has been a continuous challenge. Several spectroscopic techniques, among which atomic absorption spectroscopy (AAS) and inductively coupled plasma mass spectrometry (ICP-MS), have been first employed for trace level analysis of heavy metals.<sup>5,6</sup> However, electrochemical techniques have been found to be more attractive in this area as they provide high analytical performance (in terms of selectivity, precision, and accuracy) at a much lower cost compared with spectroscopic techniques, using portable instrumentation which allows *in situ* analysis.<sup>7–10</sup>

Voltammetric methods, especially stripping techniques, mainly anodic stripping voltammetry (ASV) combined with square-wave and differential pulse excitation signals *i.e.*, SWASV and DPASV, respectively, have very low detection limits.<sup>11</sup> In the first step of ASV (the pre-concentration step), the metal ions are electrodeposited (reduced) from the solution on the surface of the electrode at an applied potential. In the second step (stripping step), the potential is swept in the anodic direction, and at a specific potential, the electrodeposited metal is oxidized and stripped out of the electrode into the solution. The current flowing in this step is proportional to the concentration of the metal present.

Different types of mercury electrodes (*i.e.*, hanging mercury drop electrodes, dropping mercury electrodes, and mercury film electrodes) have been first employed in trace metal analysis.<sup>7,9,12,13</sup> Although the mercury electrodes have, among others, high sensitivity and reproducibility, due to mercury's toxicity and its ability to accumulate, they are no longer desirable for this determination.7,14 Glassy carbon electrodes (GCEs) and carbon paste electrodes (CPEs) have been extensively employed in the determination of several heavy metals in various environments. In addition to the most common electrodes mentioned above, several other approaches have also been used to develop electrodes for trace metal analysis, including 3D printed electrodes and electrochemical paperbased analytical devices (ePADs).<sup>15-20</sup> The ease of fabrication onto paper combined with the possibility of performing highly selective determination with very low detection limits of target analytes, using miniaturized electrodes, renders ePADs very attractive in trace metal analysis under both static and hydrodynamic conditions.<sup>17,18,20,21</sup> In addition, these devices are non-expensive and easily disposable.<sup>20</sup> Screen-printing technology is one of the most common methods for ePAD manufacturing, with screen-printed electrodes (SPEs) being among the most used electrodes. 3D printing technology allows the low cost, in-house manufacturing of highly reproducible electrodes with customizable geometries using a wide range of materials.<sup>15,16,19,22</sup> Photopolymerization, extrusion, powder bed fusion and material jetting are the most used methods for the fabrication of 3D printed electrodes.15,16 In order to increase charge transfer and to obtain reproducible active surfaces, different types of surface activations are sought for 3D printed electrodes, including metal deposition, and chemical and electrochemical treatments.<sup>16</sup>



**Fig. 1** Distribution of the research work (presented in this review) published in the last five years (*i.e.*, 2018–2022, including the research work published so far in 2023) on modifications of electrodes used in trace heavy metal analysis based on (a) the year of publication and (b) the type of electrode modifier used.

However, independent of their type, in order to improve the sensitivity and/or selectivity of all types of electrodes, further modifications are always sought. As shown in Fig. 1b, a wide range of different materials and/or compounds, including metallic and polymeric film-forming substances, various types of nanomaterials (*i.e.*, nanoparticles (NPs), nanotubes, nanocomposites, and nanosheets), metal- and covalent–organic frameworks, polymers, and environmentally friendly modifiers (*i.e.*, biopolymers, plant extracts, natural products, *etc.*) have been employed.

In the following sections, the main materials and/or compounds are discussed (grouped based on their type), along with the explanation of their use in combination with other modifiers to obtain the modified electrodes for trace heavy metal analysis. This review will first discuss the modification of electrodes using different types of nanomaterials, followed by film-formation. Next, the role of polymers, metal- and covalent-organic frameworks, other organic compounds, as well as environmentally friendly compounds, and natural products as electrode modifiers will be discussed. In addition, for each of the main groups of the modifiers mentioned above, the main analytical performance parameters of the modified electrodes (including the limit of detection (LOD) and linear concentration range) and the respective testing conditions, as well as possible interferences affecting their sensitivity are presented in Tables S1-S6 in the ESI.† The application of the modified electrodes for trace metal analysis in real samples will also be discussed.

# 2. Nanomaterials as electrode modifiers

The combination of a large specific surface area and high conductivity renders nanomaterials (NMs) very attractive as electrode modifiers in trace heavy metal analysis. Five main types of NMs, i.e., graphene, graphene oxide (GO) and reduced graphene oxide (rGO),<sup>22–92</sup> NPs,<sup>23–25,27,31,56,57,59,64,67,70,83,86,87,93–138</sup> single and (SWCNTs multiwalled carbon nanotubes and MWCNTs),<sup>32,83,89,98,99,101,113,129,133,139–162</sup> nanosheets,<sup>34,85,107,111,139,162–166</sup> and nanofibers,<sup>167–169</sup> have been employed in the modification of the electrodes used in the determination of heavy metals. Nanocubes, nanoflakes, nanospheres, and nanodiamonds  $^{75,170-175}$  are among the other types of NMs used in the modification of the electrodes. Moreover, a poly(melamine)/grafitic-C<sub>3</sub>N<sub>4</sub> nanonetwork,<sup>176</sup> NiCo<sub>2</sub>O<sub>4</sub> nanocages,<sup>177</sup> Co<sub>3</sub>O<sub>4</sub> nanopolyhedrons,<sup>88</sup> and N,S-dual-doped carbon/sepiolite clay hybrid nanostructures<sup>178</sup> were also used in the modification of various types of electrodes.

NMs have been primarily applied on the electrodes in combination with other modifiers (including through the formation of composites).<sup>22–25,27,30–32,34,46,47,50,54,59,67,70,72,75,83,85,86,88,89,92–102,107,</sup> <sup>113,115,117,118,122–126,129–131,135,136,138–156,160,162,168,175,179–189</sup> However,

in most of these studies, due to the lack of discussing the sensing mechanism of these nanocomposite-modified electrodes, there is no clear understanding of the influence of different components on the sensitivity and/or selectivity of the electrodes. Apart from the combination of different NMs (usually a combination of NPs and MWCNTs),<sup>83,98,99,101,107,113,115,128,129,133,139,162</sup> compounds and/or materials belonging to the other main groups of electrode modifiers have also been combined with NMs to modify the electrodes. As a result, many of the studies reported on the use of nanocomposites are further discussed in sections 3–7. Several studies also reported on the use of NMs as sole electrode modifiers.<sup>105,109,112,114,119,121,127,134,137,161,163,165,166,170,172,173,190</sup>

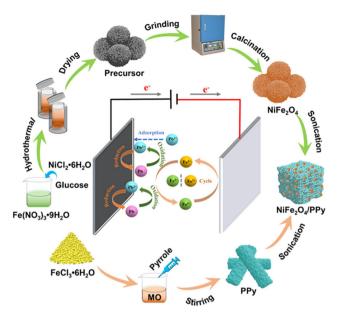
Table S1 in the ESI† summarizes the main findings of the research studies published in the last five years on using different NMs to determine trace heavy metals. The parameters related to the analytical performance of the modified electrodes with composites comprised of other main group modifiers are presented in Tables S2–S6 in the ESI.†

Carbon-based nanomaterials, including graphene, GO, rGO, carbon nanotubes (CNTs), carbon nanofibers, and carbon nanosheets, are widely used in the modification of electrodes. In addition to the above-mentioned properties, carbon-based NMs are easy to apply, low cost, and abundant materials which can be further functionalized and serve as hosts for other compounds/ materials (mainly NPs).<sup>191,192</sup> These materials are mostly used either in combination with other modifiers,<sup>24,30,34,47,54,56,57,59,64,67,70,75,85,86,88,107,113,115,129,147,149,160,162</sup> or functionalized with other compounds.<sup>146,148,158,159,167</sup>

In general, organic functionalization (i.e., oxygen and amino functionalization) and doping (mainly with nitrogen, boron, sulphur, and phosphorus) are the two main strategies used to improve the catalytic activity and increase active sites (affecting in this manner the electron-transfer process) and the hydrophilic nature of the carbon NMs.<sup>73,85,147,193,194</sup> Co-doping rGO with either nitrogen and sulphur<sup>67,178</sup> or nitrogen and bismuth<sup>195</sup> has also been reported. In order to overcome the insolubility and their tendency to agglomerate, as well as to improve their biocompatibility, CNTs are subjected to covalent (acid and functionalization treatment) non-covalent functionalization by polymers, surfactants, or aromatic compounds.<sup>32,98,101,142,148,154,158</sup> However, the use of CNTs also faces challenges related to their synthesis (i.e., cost and specific route) as well as the need for binders to improve their mechanical stability.196

Metals (*i.e.*, Ag, Au, Bi, Pd, Pt, Pb, and Sn),<sup>23–25,56</sup>, 57,59,64,70,83,86,87,93–95,97–99,101,102,104–106,108,110,113–115,117,120,121,123,

<sup>125–127,129,132,134,138</sup> metal oxides and mixed metal oxides (*i.e.*, SiO<sub>2</sub>, SnO<sub>2</sub>, Fe<sub>3</sub>O<sub>4</sub>, Mn<sub>3</sub>O<sub>4</sub>, Cu<sub>2</sub>O/ZnO, FeAl<sub>2</sub>O<sub>4</sub>, MgFe<sub>2</sub>O<sub>4</sub>, ZnFe<sub>2</sub>O<sub>4</sub>, and NiFe<sub>2</sub>O<sub>4</sub>)<sup>27,31,67,96,100,103,109,112,118,122,124,130, 131,133,135–137,188,197,198</sup> are the most commonly used NPs in the modification of the electrodes. In addition, clinoptilolite (a naturally occurring zeolite containing Si and Al) NPs were ion exchanged in a Fe( $\pi$ ) solution, and the modified CPE obtained was used in the determination of Sn<sup>2+, 199</sup> Several research groups have also reported on the synthesis of magnetic NMs (mainly NPs as part of nanocomposites) and their use as electrode modifiers.<sup>26,30,96,100,132,137,182,188,200</sup> Fig. 2 presents the preparation of magnetic NiFe<sub>2</sub>O<sub>4</sub>-NPs and polypyrrole (PPy) nanocomposites used in the modification of a GCE for the determination of Pb<sup>2+</sup>



**Fig. 2** Schematic illustration of the preparation process of the NiFe<sub>2</sub>O<sub>4</sub>/ PPy nanocomposite.<sup>96</sup> Reprinted from Y. Wang, Z. Nie, X. Li, R. Wang, Y. Zhao, and H. Wang, PPy-functionalized NiFe<sub>2</sub>O<sub>4</sub> nanocomposites toward highly selective Pb<sup>2+</sup> electrochemical sensing, *ACS Sustainable Chem. Eng.*, **10**(18), 6082–6093, Copyright (2022) American Chemical Society.

in tap, lake, and river water samples.<sup>96</sup> Although the performance of the NM-modified electrodes has been mainly tested in real water samples, several studies reported on their use in heavy metal determination in other samples, including, among others, body fluids<sup>31,57,88,163,173,190</sup> and cosmetic product<sup>190</sup> samples. Moreover, Das *et al.*<sup>31</sup> successfully employed a bimetal oxide ( $Bi_2O_3/Fe_2O_3$ )-NP decorated GO modified PGE for the determination of Cd<sup>2+</sup> in a wide range of real samples, including food (potato, lemon, and apple), soil, biological samples (blood serum and urine), and water (tap, drinking and chemical laboratory wastewater) samples.

In addition to the widely used electrodeposition, reduction, and hydrothermal methods, the synthesis of NPs from environmentally friendly sources (*i.e.*, waste and plant extracts as well as mango seed kernels) has been reported,<sup>23,119–122,125,127</sup> and will be further discussed in section 7.

### 3. Polymers as electrode modifiers

Polymers' high conductivity and porosity, combined with the presence of several functional groups and their ability to be further functionalized, make polymer-modified electrodes very attractive in trace heavy metal determination. Table S2 in the ESI† summarizes the main findings of the research studies published in the last five years reporting on the use of different polymer modifiers for various types of electrodes employed in the determination of trace heavy metals.

Although the vast majority of the studies reported on the combined use of polymers and other materials and/or mole-

cules to modify the electrodes,  $^{22,23,26,29,32,35,37,38,41,45,61,69,}_{71-73,77,80,84,87,89,91,92,94,96-102,106,122-124,131,141-144,175,176,182-184,}$ 

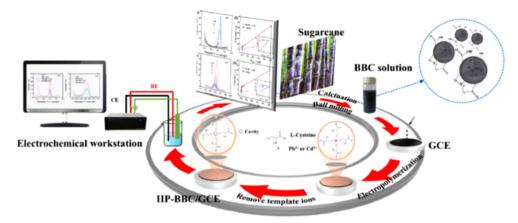
<sup>186,187,197,200–221</sup> there is also a significant number of studies on polymers as sole modifiers.<sup>174,185,222–232</sup> These exceptions include, among others, the pure polymer film-modified electrodes discussed in section 4. A combination of two polymers (*i.e.*, conductive polydopamine (PDA) doped PPy hydrogel) as modifiers of an SPCE for the determination of Pb<sup>2+</sup> has also been reported.<sup>232</sup>

Biopolymers (mainly chitosan and different forms of cellulose)<sup>35,37,61,69,71–73,87,89,92,123,124,175,186,187,197,200–205</sup> (further discussed in section 7) and Nafion (a sulfonated tetrafluoroethylene-based fluoropolymer–copolymer)<sup>24,62,108, 113,149,154,155,179,200,233–241</sup> are by far most frequently used as polymeric components of the nanocomposites. Moreover, other conducting polymers such as polyaniline (PANI),<sup>23,38,99,102,122,144,183,206–209,220</sup> PPy,<sup>32,77,80,96,101,</sup>

 $^{(112,2)}_{^{210-212,214,216}}$  and poly(3,4-ethylenedioxythiophene) (PEDOT) derivatives,  $^{58,174,217}$  as well as PDA,  $^{98,182}$  have also been part of the nanocomposites used to modify electrodes for trace heavy metal analysis. In addition, nanocomposites comprised of ion imprinted polymers (IIPs)<sup>29,41,84,87,100,187,214,219</sup> as well as ion imprinted NPs<sup>206,215</sup> have also been reported as modifiers. Fig. 3 shows the preparation steps for an ion imprinted (L-cysteine) and ball milling biochar (BBC) obtained from the high temperature pyrolysis of sugarcane-modified GCE used in the individual determination of Pb<sup>2+</sup> and Cd<sup>2+</sup> in tap, lake, and ground water samples.<sup>219</sup> The authors reported that low LOD values of 5.86 fM and 0.883 aM for Pb<sup>2+</sup> and Cd<sup>2+</sup>, respectively, were achieved with the modified electrode.

Nandy et al.97 synthesized a block copolymer from poly (methyl methacrylate) and poly(2-acrylamido-2-methylpropane sulfonic acid) and, in combination with Ag-NPs used it to modify a GCE for the determination of Hg<sup>2+</sup>. Different forms of carbon-containing materials (i.e., graphene, GO, rGO, CNTs, and  $g-C_3N_4$ ,  $2^{29,32,38,45,80,87,91,98,99,101,141,142,144,176,183,184,206}$  NPs nanoflakes,<sup>23,87,94,96–99,101,102,124,176,182,200,218</sup> and and MOFs<sup>208,209,213,216</sup> were combined with the polymers and biopolymers to obtain the modified electrodes. Chelating agents, such as 2,2',2",2"'-(ethane-1,2-diyldinitrilo)tetraacetic acid (EDTA) and L-cysteine, have also been included in these polymer-containing nanocomposites.<sup>207,214,220</sup> Meanwhile, Fernando et al.<sup>142</sup> reported on the combination of a chelating polymer (i.e., poly(4-vinyl pyridine)) with MWCNTs and poly (ethylene terephthalate) as modifiers of an Au electrode for Cu<sup>2+</sup> determination in tap, lake, and ocean water samples. Apart from different types of real water samples, few studies reported on polymer-modified electrodes for heavy metal determination in food and beverages<sup>100,185,242</sup> and body fluid samples.<sup>106,185,209</sup>

*In situ* electrochemical and oxidative polymerization, <sup>38,80,174,176,211,214,219</sup> as well as co-precipitation polymerization, <sup>26,100,215</sup> are among the methods used to prepare the nanocomposites. Suspensions of these nanocomposites in different solvents (including ethanol, isopropanol, *N*,*N*-dimethylformamide, and water) were dropcast on

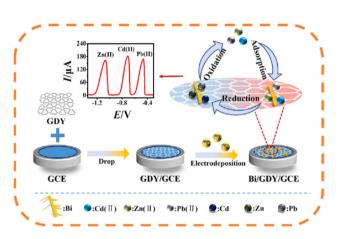


**Fig. 3** Schematic diagram for the preparation of the ion imprinted BBC electrode (IIP-BBC/GCE) and application for the detection of heavy metal ions<sup>219</sup> (this figure was published in D. Mao, J. Hu, P. Duan, C. Qin, Y. Piao, Ultrasensitive and highly reusable electrochemical sensor with ion imprinted nanobiochar, *Sens. Actuators, B*, **371**, 132490, Copyright Elsevier (2022)).

the surface of the electrodes. In the case of modified CPEs, the polymers were mixed with graphite and other additives to form the paste.

# 4. Film-forming substances as electrode modifiers

The development of film-coated electrodes presented an excellent alternative to the mercury electrodes in trace heavy metal analysis.7,243 Since the first Bi-coated carbon electrode proposed by Wang et al.,<sup>244</sup> several mainly metallic (including Sb-, Sn-, Pb-, and Cu-film electrodes) and other film-coated electrodes have been employed in the determination of various heavy metals.<sup>7,12,243</sup> In the last five years, the research was focused on preparing two main types of film electrodes, *i.e.*, metallic-<sup>25,40,50,83,181,197,233,235,245-273</sup> and polymeric-film electrodes.<sup>77,91,94,95,141,210,217,220,223-230,274-276</sup> However, the combination of metallic (Bi) and polymer films (i.e., PPy and aminonaphthalenesulphonic acid derivatives), in addition to other materials, was also used to modify GCEs for the determination of Pb<sup>2+</sup> and Cd<sup>2+</sup>.<sup>216,221</sup> Moreover, the development of a bimetallic (Hg-Bi)-film incorporated with a poly(1,2-diaminoanthraquinone) modified GCE for individual and simultaneous determination of Pb<sup>2+</sup>, Cd<sup>2+</sup>, and Zn<sup>2+</sup> was reported.<sup>277</sup> Oghli and Soleymanpour<sup>218</sup> reported on combining poly-Lcysteine (a conducting polymer) and a Fe<sub>3</sub>O<sub>4</sub>-NPs film to modify a pencil graphite electrode (PGE) for the determination of Hg<sup>2+</sup>, Cu<sup>2+</sup>, and Cd<sup>2+</sup>. Furthermore, carbon and several of its forms (*i.e.*, diamond, graphdiyne, rGO, and g-C<sub>3</sub>N<sub>4</sub>) have also been coated on different types of film electrodes.<sup>76,83,93,139,246,278</sup> Fig. 4 presents the formation of a dendritic bismuth and graphdiyne (Bi/GDY) film on the surface of a GCE used for the simultaneous determination of Zn<sup>2+</sup>, Cd<sup>2+</sup>, and Pb<sup>2+</sup> in real seawater samples.<sup>246</sup> The authors attributed the low LODs obtained (i.e., 0.115 nM, 0.171 nM, and 0.146 nM for Zn<sup>2+</sup>, Cd<sup>2+</sup>, and Pb<sup>2+</sup>, respectively) to the



**Fig. 4** The preparation of the Bi/GDY/GCE and the application for simultaneous detection of three heavy metal ions<sup>246</sup> (this figure was published in Y. Ai, L. Yan, S. Zhang, X. Ye, Y. Xuan, S. He, X. Wang, W. Sun, Ultra-sensitive simultaneous electrochemical detection of Zn(n), Cd(n) and Pb(n) based on the bismuth and graphdyne film modified electrode, *Microchem. J.*, **184**, 108186, Copyright Elsevier (2023)).

synergistic effect of the Bi-film and the GDY. Table S3 in the ESI<sup>†</sup> summarizes the main findings of the research studies published in the last five years regarding the use of different types of film electrodes in the determination of trace heavy metals.

Bismuth film electrodes (BiFEs) are among the most employed electrodes in the determination of trace heavy metals.<sup>25,40,50,83,181,197,233,235,245-260</sup> In addition to its low toxicity and the ability to form alloys with other metals, bismuth has a wide potential range, which allows BiFEs to have comparable analytical performance to the mercury electrodes in trace heavy metal analysis.<sup>7,243,244</sup> The film can be electrochemically deposited directly on the electrode surface from the solution containing both the modifier and the analytes (*in situ* preparation) or through film deposition before the electrode is introduced in the sample solution (*ex situ* preparation). In the

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studies published in the last five years, bismuth was almost always in situ electrodeposited on the electrode surface, with only a few exceptions.<sup>197,248</sup> Trace heavy metal analysis has been performed either with just BiFEs<sup>247,248,252,253,255-257,260</sup> or with electrodes modified with a Bi-film combined with other materials and/or compounds, including with different NMs,<sup>25,50,83,181,256</sup> Nafion and GO,<sup>40,83,233,235,250</sup> as well as doping of the Bi-films with boron or indium.<sup>245,259</sup> Various other metals, including Sb,<sup>261-264</sup> Cu,<sup>265-267</sup> Au,<sup>268</sup> Pb,<sup>269</sup> and Hg,<sup>260</sup> have also been electrodeposited on different types of electrodes. Moreover, complexing agents such as 8-hydroxyquinoline,<sup>251</sup> nioxime,<sup>265</sup> and cupferron<sup>269</sup> have been added during the in situ Bi-, Pb-, and Cu-film modification of GCEs. Next, the combination of two or three metals such as Bi-Sn, Bi-Cu, Bi-Hg, and Bi-Sn-Sb has also been reported for the modification of GCEs used in the determination of Pb<sup>2+</sup>, Cd<sup>2+</sup>, and  $Zn^{2+}$ .<sup>270–272,277</sup> The main advantage of these types of electrodes is their increased sensitivity compared with single metal film electrodes. Different Bi: Cu, Bi: Sn, and Bi: Sn: Sb ratios (for a total mass concentration of either 0.5 or 1.0 mg L<sup>-1</sup>) have been employed to in situ modify GCEs.<sup>270-272</sup> No direct correlation between the Bi to metal ratio and the LOD or linear concentration range of all the analytes of interest has been observed in these studies.

Polymer film electrodes are obtained primarily by electropolymerization of the monomers on the electrode surface<sup>94,141,223-230,274,275</sup> However, Yi *et al.*<sup>91</sup> drop-coated the surface of the GCE with a suspension of poly(L-glutamic acid) and GO in *N*,*N*-dimethylformamide, and used the modified film electrode for simultaneous determination of  $Cu^{2+}$ ,  $Cd^{2+}$ , and  $Hg^{2+}$ . Aminoanthraquinone<sup>141,229,230,274</sup> and PPy derivative polymers,<sup>77,210,216,223</sup> are among the most used conductive polymers to coat the electrodes. The modified electrodes are obtained either by pure polymer film-formation,<sup>223–230</sup> or by combining the polymers with other materials and/or compounds.<sup>91,94,95,141,210,220,274,276</sup>

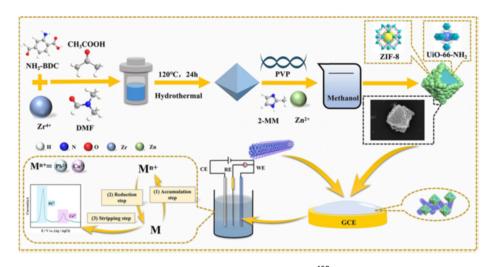
# 5. Metal- and covalent-organic frameworks as electrode modifiers

Table S4 in the ESI<sup>†</sup> summarizes the main findings of the research studies published in the last five years regarding the use of different metal-organic frameworks (MOFs) and covalent-organic frameworks (COFs) as modifiers for various types of electrodes employed in the determination of trace MOFs<sup>42,53,116,150-154,164,177,</sup> heavy metals. Several <sup>201,208,209,212,216,236,237,279-297</sup> and COFs<sup>198,298-302</sup> have been employed as modifiers for different types of electrodes in combination with other materials. MOFs are 1D, 2D, or 3D network like structures, highly ordered crystalline materials formed by self-assembly of metal ions or clusters and organic ligands through coordination bonds.<sup>303</sup> A similar type of structure is observed in the case of COFs, but instead of the metal COFs they contain light elements (such as C, H, O, N, or B atoms) connected with the rest with strong covalent bonds.<sup>304</sup> Both types of materials possess a large specific surface area, high and tunable porosity, and, most importantly, can be easily functionalized or combined with other materials. These properties make these materials suitable for applications in several areas, including as modifiers for different types of electrodes.<sup>303,304</sup>

However, two of the main drawbacks of MOFs and COFs in trace heavy metal determination are their low conductivity and low stability in water. To increase the charge transfer (*i.e.*, to facilitate the electron transfer through the electrode surface to or from the electroactive species in solution) and to improve their mechanical strength, MOFs have been combined with carbon-containing materials, such as graphene (mainly rGO),<sup>42,53,285</sup> MWCNTs,<sup>150,152-154</sup> mesoporous carbon,<sup>282</sup> carbon black,<sup>236</sup> and expanded graphite.<sup>286</sup> The addition of conducting polymers (mainly PANI and PPy)<sup>208,209,212,213,216</sup> to MOFs has resulted in higher conductivity and stability in water. The development of core-shell structures has been reported to not only increase the stability, but also improve the LOD of the modified electrodes used for the determination of Pb<sup>2+</sup>, Cd<sup>2+</sup>, and Cu<sup>2+</sup> ions.<sup>150,209</sup> These structures are formed through either the integration of two MOFs (*i.e.*, NH<sub>2</sub>-UiO-66 and ZIF-8 to form NU66@Z8, shown in Fig. 5),<sup>150</sup> or the combination of a MOF (*i.e.*, NH<sub>2</sub>-UiO-66) and self-doped PANI nanofibers.<sup>209</sup> The functionalized NU66@Z8 (shown in Fig. 5) was applied on the GCE and the modified electrode was used in the simultaneous determination of Pb<sup>2+</sup> and Cu<sup>2+</sup> in tap water of laboratory, soil, and Chinese cabbage samples.<sup>150</sup>

Moreover, the integration of Bi in the form of films or encapsulated in the mesoporous structure of MOFs and COFs, has also been reported.<sup>198,216,295</sup> Liu *et al.*<sup>216</sup> and Sarvestani *et al.*<sup>198</sup> used an *in situ* formed Bi-film and MOFs or COFs to modify a GCE surface for the determination of Pb<sup>2+</sup>. Few studies reported on the use of newly synthesized MOFs and COFs as sole electrode modifiers.<sup>284,288,291,302</sup>

Zeolitic imidazolate frameworks (ZIFs),<sup>154,177,201,236,279,284,286</sup> amino-functionalized Zr-based (Universitet i Oslo MOFs (UiO) type, *i.e.*, NH<sub>2</sub>-UiO-66),<sup>42,53,151,152,209,216,283,285</sup> and Materials of Institute Lavoisier (MILs),<sup>164,212,295</sup> are among the main types of MOFs used as electrode modifiers. The presence of amino groups or imidazole in MOFs facilitates the formation of coordination bonds with heavy metals, improving the adsorption ability of the analytes and thus improving the analytical performance of the electrodes. The majority of COFs used were prepared by amine-aldehyde condensation. The metals incorporated in the MOF structures include Zr, Zn, Cu, Cr, Bi, Fe, V, Co, Mn, Ca, and lanthanides. However, bimetallic ZIF-type MOFs (i.e., Zn and Ni<sup>236</sup> and Fe and Co<sup>284</sup>) have also been reported as modifiers for GCEs. Moreover, ZIF-8 modified with mesoporous carbons served as the substrate for the in situ growth of MIL-100(Fe), and the obtained hybrid material from the combination of two MOFs was used to modify the GCE for the simultaneous determination of Cd<sup>2+</sup> and Pb<sup>2+</sup>.<sup>282</sup> In addition, MOFs (i.e., ZIF-67) can serve as precursors for the synthesis of 3D structures, such as NiCo2O4 nanocages, employed to modify the GCE, enabling Hg<sup>2+</sup> determination.<sup>177</sup>



**Fig. 5** The synthesis route of NU66@Z8 and the construction of the working electrode<sup>150</sup> (this figure was published in R. Tan, P. Jiang, C. Pan, J. Pan, N. Gao, Z. Cai, F. Wu, G. Chang, A. Xie, Y. He, Core–shell architectured  $NH_2$ -UiO-66@ZIF-8/multi-walled carbon nanotubes nanocomposite-based sensitive electrochemical sensor towards simultaneous determination of Pb<sup>2+</sup> and Cu<sup>2+</sup>, *Microchim. Acta*, **190**, 30, Copyright Springer (2023)).

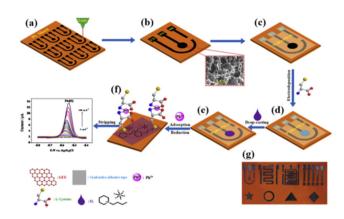
Dropcasting dispersions of the MOFs in various solvents (including ethanol, N,N-dimethylformamide, and isopropanol) on the pretreated surfaces of the electrodes, followed by drying at room temperatures, is generally the main mechanism for applying these frameworks. Although GCEs are by far the most commonly used substrates for the application of different COFs, 53,116,150,152-154,177,198, types of MOFs and 201,216,236,237,279-290,298,299 several other types of electrodes have also been modified, including CPEs, 291,292,301 SPCEs, 42,209 gold electrodes, 212,293 as well as PGEs, SPEs, and carbon cloth electrodes (CCEs).<sup>208,294,295,298</sup> In addition, Yang et al.<sup>296</sup> and Han et al.<sup>300</sup> reported on the attachment of a mercaptan functionalized Zr-based MOF as well as a 1,4-benzenedithiol-2,5-diaminohydrochloride and 1,3,5-triformylbenzene condensed COF to a 3D-structure kenaf stem derived carbon. These combined electrodes have been employed in either the simultaneous determination of  $Cd^{2+}$ ,  $Pb^{2+}$ ,  $Cu^{2+}$ , and  $Hg^{2+}$  or for the determination of Hg<sup>2+</sup> individually. The authors reported that the integrated electrode has better analytical performance compared with the GCE modified electrode.<sup>296</sup> In addition to different real water samples, modified electrodes have also been successfully employed in the determination of heavy metals in food, 42,53,116,151,236,293 spices, 198,302 and body fluid 208,209 samples.

# 6. Other organic compounds as electrode modifiers

In addition to the above-mentioned main groups of materials and/or compounds, in the last five years, the research community has also reported on the inclusion of calixarene-based compounds,<sup>117,118,281,305-309</sup> ionic liquids (ILs),<sup>55,155,168,258</sup> and Schiff bases<sup>156,310,311</sup> as electrode modifiers for trace heavy metal analysis. Table S5 in the ESI† summarizes the main findings of the research studies published in the last five years reporting on the use of other organic compounds as modifiers for various types of electrodes employed in the determination of trace heavy metals.

Calix[n]arenes are macrocyclic compounds containing several phenolic units (*n* represents the number of phenolic units).<sup>312</sup> Their cavity-containing, cup-shaped structure with both polar and nonpolar groups, combined with the ability to form complexes with cations, in addition to their ease of synthesis and functionalization, makes these compounds attractive in trace metal analysis. In the last five years calix[4]arenefunctionalized compounds have been used mainly as sole electrode modifiers. However, their use in combination with NPs (*i.e.*, Au- and  $Mn_3O_4$ -NPs),<sup>117,118</sup> as well as with a MOF<sup>281</sup> was also reported. Calix[4]arene-functionalized compounds were mostly used to modify GCEs (in addition to the SPCE and Au electrode) employed in the individual or simultaneous determination of Hg<sup>2+</sup>, Pb<sup>2+</sup>, Cd<sup>2+</sup>, Zn<sup>2+</sup>, and Cu<sup>2+</sup> in different real water samples (including wastewater, river, drinking, and deionized water)<sup>117,118,305-307,309</sup> and commercial oysters, clams, and mussel samples.<sup>308</sup>

Imidazolium-<sup>155,168</sup> and pyridinium-based<sup>55,258</sup> ILs are also among the organic compounds used to modify the electrodes. These ILs were always applied in combination with other materials and/or compounds (*i.e.*, nanofibers, SWCNTs, Bi and Fe<sub>3</sub>O<sub>4</sub>-NPs, and L-cysteine). Apart from the conventional electrodes (GCE, SPE, and CPE), a pyridinium-based IL (*i.e.*, *N*-butylpyridinium hexafluorophosphate) in combination with poly-L-cysteine has been used to modify a laser-engraved electrode (LEGE) as shown in Fig. 6. The authors have attributed the increased sensitivity of the modified LEGE to the combination of the good conductivity of the IL and the complexing ability of poly-L-cysteine. The modified electrodes have been successfully employed in the individual or simultaneous determination of Pb<sup>2+</sup> and Cd<sup>2+</sup> in real water<sup>55,168</sup> and soil extract<sup>155,258</sup> samples. **Critical Review** 



**Fig. 6** Schematic of the fabrication process of LEG micro-patterned electrodes for the electrochemical sensing of lead ions. (a) Fabrication of LEGE arrays on a PI sheet by direct laser engraving. (b) Three-electrode system of the LEGE. (c) Selective surface passivation of LEGE areas by a parafilm to define the WE area. (d) L-Cysteine was polymerized on the electrode to form the PLC/LEGE. (e) IL-functionalized PLC/LEGE. (f) Selective electrochemical detection of Pb<sup>2+</sup>. (g) Optical image of different micro-patterned electrode arrays on a PI sheet<sup>55</sup> (this figure was published in Z. Lu, X. Lin, J. Zhang, W. Dai, B. Liu, G. Mo, J. Ye, J. Ye, lonic liquid/poly-L-cysteine composite deposited on flexible and hierarchical porous laser-engraved graphene electrode for high-performance electrochemical analysis of lead ion, *Electrochim. Acta*, **295**, 514–523, Copyright Elsevier (2019)).

The ability of the Schiff bases to form complexes with metal ions through the lone pair electrons of O and N atoms in their structure makes this type of compound of interest potent in the modification of electrodes for trace metal analysis. Schiff bases have been used either as sole modifiers<sup>310,311</sup> or applied in combination with MWCNTs<sup>156</sup> on the surface of CPEs. The modified electrodes have been successfully employed mostly in the determination of Pb<sup>2+</sup> and Hg<sup>2+</sup> in diverse samples, including real water,<sup>310,311</sup> food,<sup>310</sup> tobacco,<sup>310,311</sup> and algae<sup>156</sup> samples.

# 7. Environmentally friendly compounds and natural products as electrode modifiers

In the last five years, several research groups have continued to explore the inclusion of various environmentally friendly (green) modifiers in the development of new electrodes for trace metal analysis.<sup>23,35,37,61,66,69,71–73,87,89,92,104,110,120–127, 157,175,186,187,197,201–205,219,289,293,313–321</sup> Table S6 in the ESI† summarizes the main findings of the research studies published in the last five years reporting on the use of different green modifiers for various types of electrodes employed in the determination of trace heavy metals.

A wide range of green modifiers has been employed, including biopolymers,  $^{35,37,61,69,71-73,87,89,92,123,124,175,186,187,197,201-205}$  plant extracts,  $^{23,66,104,120-122,125,127,313}$  natural products,  $^{126,219,314-318}$  biomass,  $^{157,289,319}$  and DNA. $^{293,320,321}$  It

is important to emphasize that all the different green modifiers have been either added to or combined with other modifiers to achieve the desired electroanalytical performance of the newly developed electrodes through the synergistic effect. Various other modifiers (discussed in sections 2–6) have been combined with the green modifiers to obtain the final modified electrodes. Moreover, Wong *et al.*<sup>175</sup> reported on combining two green modifiers, chitosan, and biochar (*i.e.*, carbon NPs with a large surface area and temperature dependent hydrophobicity), which in addition to nanodiamonds were used to modify a GCE for Pb<sup>2+</sup> and Cd<sup>2+</sup> determination in river water samples. Low LODs (*i.e.*, 0.056  $\mu$ M and 0.110  $\mu$ M for Pb<sup>2+</sup> and Cd<sup>2+</sup>, respectively) were achieved with the modified electrode.<sup>175</sup>

In addition to the above-mentioned green modifiers, few research groups reported on environmentally friendly approaches to obtain the modified electrodes. The determination of  $Pb^{2+}$  and  $Cd^{2+}$  in either various vegetables<sup>65</sup> or herbal food supplements<sup>239</sup> was carried out with a multilayer graphene paste electrode modified with activated carbon produced from coconut husk or using the graphite extracted from waste zinc-carbon batteries combined with Bi-NPs, MWCNTs, and Nafion, respectively. Pristine blast furnace  $slag^{322}$  has also been used to modify a GCE for the determination of  $Pb^{2+}$  and  $Cu^{2+}$ . However, the determination of 1 mM of the analytes was significantly influenced by the presence of 2 mM Cd<sup>2+</sup>, Zn<sup>2+</sup>, and Hg<sup>2+</sup>.

Biopolymers are by far the most used green modifiers for the various types of electrodes used in trace heavy metal analysis. This is due to the fact that apart from being biocompatible, biodegradable, nontoxic, and relatively inexpensive, they combine high mechanical strength and water permeability, good adherence to surfaces, good film-forming ability, and high hydrophilicity with the ability to form chelates with transition metal ions.<sup>123</sup> These compounds are rich in functional groups such as hydroxyl, carboxyl, and sulfonic groups, which bind the analytes and help facilitate the combination of modifiers.<sup>69,71,72,92</sup> Chitosan,<sup>35,37,61,73,87,</sup> different <sup>89,123,124,175,186,187,197,201</sup> modified cellulose, bacterial cellulose and carboxymethyl cellulose (CMC),<sup>69,71,202,203</sup> lignosulfonate,<sup>92</sup> chitin,<sup>204</sup> lignin,<sup>205</sup> and  $\kappa$ -carrageenan<sup>72</sup> are among the biopolymers used to modify the electrodes.

In most studies, plant extracts have been used as precursors of NPs (mainly Ag-NPs)<sup>23,104,120,121,125,127</sup> to modify electrodes for the determination of Pb<sup>2+</sup>, Cd<sup>2+</sup>, and Hg<sup>2+</sup> ions. The synthesis and characteristics of Ag-NPs, as well as the performance of the obtained Ag-NPs modified electrode, were found to be pH dependent.<sup>127</sup> Okpara *et al.*<sup>122</sup> synthesized metal oxide NPs from orange and lemon peel extracts and used them to modify an SPCE to determine Cd<sup>2+</sup> and Hg<sup>2+</sup> ions. Meanwhile, the powder obtained from *Eichhornia crassipes* root extract was incorporated into a CPE and used for Pb<sup>2+</sup> and Cd<sup>2+</sup> determination.<sup>313</sup>

Natural products such as spent coffee grounds,<sup>314,315</sup> *Bigarreau Burlat* kernel shells,<sup>316</sup> walnut shells,<sup>317</sup> tapioca flour,<sup>126</sup> mango seed kernels,<sup>318</sup> sugarcane,<sup>219</sup> and waste pomelo peels<sup>110</sup> have also been used as green modifiers, with

and without pretreatment. The spent coffee grounds were first treated with citric acid to increase the carboxyl groups present, and then, in addition to other modifiers, were used to prepare the modified CPE employed for the determination of  $Cd^{2+}$  and  $Pb^{2+,314}$  Spent coffee grounds,<sup>315</sup> walnut shells,<sup>317</sup> and sugarcane<sup>219</sup> were subjected to pyrolysis to obtain biochar, which, combined with other compounds, was used to modify GCEs or CPEs for the determination of  $Cu^{2+}$ ,  $Pb^{2+}$ ,  $Cd^{2+}$ , and  $Hg^{2+}$  ions. No prior chemical pretreatment of *Bigarreau Burlat* kernels shells<sup>316</sup> and mango seed kernels<sup>318</sup> was reported. Zhang *et al.*<sup>110</sup> obtained porous activated carbon from waste pomelo peels through a KOH activation method followed by carbonization at 700 °C. The porous activated carbon was then, in combination with Pd-NPs used to modify a GCE for the simultaneous determination of  $Cd^{2+}$ ,  $Pb^{2+}$ , and  $Cu^{2+}$ .

Dali *et al.*<sup>157</sup> modified a GCE with a mixture of functionalized CNTs and filamentous fungi (*Trichoderma asperellum*) and used it to determine  $Cd^{2+}$  and  $Pb^{2+}$ . The filamentous fungi can excrete organic acids which bind heavy metals. *Hordeum vulgare* L. dust (biomass residue released by brewing industries) activated using Na<sub>2</sub>CO<sub>3</sub> was used to modify a CPE for the determination of Cd<sup>2+</sup>, Pb<sup>2+</sup>, and Hg<sup>2+, 319</sup>

In all these studies, the authors claim the use of either environmentally friendly components or approaches for the modification of the electrodes, but no studies on the toxicity, biodegradability, and/or biocompatibility of the obtained composites used for electrode modification are presented.

### 8. Conclusions and outlook

The large number of publications reported in the last five years on modifications of electrodes for trace metal analysis emphasizes even more the continuous and growing interest in this area of research. A wide range of compounds and/or materials have been employed as modifiers, including nano-materials, metallic and polymeric film-formation substances, metal– and covalent–organic frameworks, polymers, and environmentally friendly compounds and natural products (*i.e.*, bio-polymers, plant extracts, *etc.*).

The vast majority of the studies published in the last five years reported on the use of different types of nanomaterials (mainly carbon-based nanomaterials and NPs) as electrode modifiers. Polymers (including biopolymers) have found growing interest in modifying the electrodes through film-formation, as sole modifiers, or in combination with other compounds/materials. Although film electrodes have been among the first alternatives to replace mercury electrodes for stripping analysis of heavy metals, even in the last five years, film formation has continued to be one of the most used alternatives for electrode modification. In addition to pure metallic and polymeric film-modified electrodes, there are a growing number of reports on the combination of different materials and/or compounds as composite materials.

The formation of these composites is not only seen in the case of film-modified electrodes, but also with other main

groups of modifiers discussed above. This is, of course, a desired outcome as it increases the possibility of developing new electrodes and/or leads to their improved sensitivity and analytical performance. However, many of these studies do not report on the contribution of the components of these composites to the analytical performance of the obtained modified electrode. It would be of great interest to the research community that the authors present the mechanism behind heavy metal determination with the modified electrodes. The latter is rarely discussed. Moreover, in some cases, the number of components involved combined with the procedures used to prepare the composites and/or apply them on the surface of the electrodes makes it difficult to scale the production of these electrodes. In several studies, the authors claim that the development of environmentally friendly electrodes is either due to the modifiers used or the preparation and modification procedure. However, no evaluation of toxicity has been presented in these studies. This is important, especially in the combination of several modifiers to form composites. Environmentally friendly compounds and natural products have also been used in the modification of electrodes, either included in the composite materials or as precursors to other materials (mainly NPs) used to modify the electrodes.

The modifications have been mostly applied to GCEs, followed by SPEs and CPEs. In addition, the modification of several other electrodes, including PGEs, Au electrodes, stainless steel electrodes, and indium tin oxide electrodes, has been reported. Only a few studies conducted in the last five years reported on the modification of 3D printed electrodes. This can be attributed to the pretreatment methods needed and the available materials that can be used to overcome the poor conductivity of such electrodes. In that respect, the modification of these types of electrodes presents a challenging area of research. However, due to their ease of preparation and modification, low cost, and portability, in combination with their high sensitivity and various applications in real samples, it would be of great interest for future research to focus more on the modification of SPEs and CPEs.

The modifiers' application on the electrode surface is mainly done through drop-casting. The modified electrodes have been successfully employed in the individual or simultaneous determination of heavy metals, primarily in diverse water samples (*i.e.*, tap, river, lake, sea, bay, spring, mountain, ground, pond, well, and reservoir water) as well as in wastewater, body fluids, and in food and drinks. There are only a few reports on using the obtained modified electrodes for heavy metal determination in soil samples.

The research work published in the last five years has been focused on the determination of the most common and most problematic heavy metals (*i.e.*,  $Hg^{2+}$ ,  $Pb^{2+}$ ,  $Cd^{2+}$ ,  $Cu^{2+}$ , etc.). Other analytes of interest included  $Zn^{2+}$ ,  $Cr^{2+}$ ,  $Cr^{3+}$ ,  $Cr^{6+}$ , and  $Tl^+$ . Most of these studies reported the development of electrodes for multiple ion determination (mostly the simultaneous determination of  $Pb^{2+}$  and  $Cd^{2+}$ ). However, in some cases, when the modified electrodes can be used in the determination of multiple ions, the authors reported only on the individual determination of the respective heavy metals. The simultaneous determination of several heavy metals is of great importance and is always sought, especially when analyzing complex samples.

Acetate and phosphate buffers are by far the most employed supporting electrolytes in these studies. The pH of these supporting electrolytes varied in the range of 2.0–6.6 for the acetate buffers and 3.5–12.0 for the phosphate buffers. Other solutions, including acids (*i.e.*, HCl and H<sub>2</sub>SO<sub>4</sub> in the 0.01–1 M and 0.1 or 0.5 M concentration range, respectively), Britton-Robinson buffer (mainly at pH = 2.0–4.0), and 0.01 or 0.1 M ammonia buffer solution (pH = 8.5 or 9.2), are also reported. 2-[4-(2-Hydroxyethyl)-1-piperazinyl]-ethanesulfonic acid (pH = 6.5 or 7.0) and 3-[(2-mercapto-vinyl)-hydrazono]-1,3-dihydro-indol-2-one (pH = 9–10) are among the organic compounds employed as supporting electrolytes for the simultaneous determination of Pb<sup>2+</sup>, Cd<sup>2+</sup>, Zn<sup>2+</sup>, and Cu<sup>2+</sup>.

The stability, reproducibility, and selectivity of the modified electrodes in the presence of possible interferents are important factors for their application in real sample analysis. The majority of studies do not report on electrode stability. The influence of a wide range of possible interferents on the selectivity of the modified electrodes has been studied. These interferents vary from the main cations and anions (*i.e.*,  $Zn^{2+}$ ,  $Ni^{2+}$ ,  $Co^{2+}$ ,  $Fe^{2+}$ ,  $Fe^{3+}$ ,  $Cu^{2+}$ ,  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $Mn^{2+}$ ,  $NH_4^+$ ,  $Na^+$ ,  $K^+$ , Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, and SO<sub>4</sub><sup>2-</sup>) to organic compounds (mainly surfactants) within 1-10 000 times the concentration of the analytes. Moreover, depending on the type of the sample analyzed, the possible interference of cationic and anionic surfactants (i.e., sodium dodecyl sulfate, cetyltrimethylammonium bromide, and Triton X-100) and other organic compounds (mainly citric, uric, tannic and ascorbic acids) has also been investigated. However, in the vast majority of reports, the interference effect is not studied for all the possible interferents that can be found in real sample analysis other than in certified reference materials. The most commonly reported interference is from Cu<sup>2+</sup> and Hg<sup>2+</sup> in the determination of Pb<sup>2+</sup> and Cd<sup>2+</sup>, respectively. The addition of ferrocyanide decreased the interference effect of the Cu<sup>2+</sup> ions.

Frequently, the authors report on one-by-one optimization of the experimental parameters. The problem with such optimization (often first the accumulation time and then the deposition time, followed by the solution composition) is that the optimum conditions in most cases cannot be met. The determination of the optimal deposition potential at a fixed accumulation time in a certain solution would result in the local improvement of e.g., sensitivity. However, when the authors fix the so-called optimized deposition potential, the other parameters change. As a result, this procedure does not lead to the optimum conditions but only to local improvements. Moreover, the authors frequently considered an increase in sensitivity as the optimization criterion. Nevertheless, the optimization of sensitivity usually leads to a narrower linear concentration range (which is of course not desired) and a different precision and accuracy of the method. The best optimization criteria should simultaneously increase the width of the linear concentration range, the sensitivity (slope of the calibration curve), the precision (the lowest RSD is desired), the highest accuracy (recovery closest to 100% is desired), and to achieve the lowest LOD and limit of quantification (LOQ) possible.

Finally, increased attention needs to be devoted to the reporting of the analytical parameters of the modified electrodes. Often authors are not consistent when reporting significant figures (number of digits in a value). For example, different numbers of digits are used to report the linear concentration range (*e.g.*, 0.1–1 nM or 31.25–2000 µg L<sup>-1</sup> or 0.02–2 ppb) and recovery (*e.g.*, 98.02–105.0% or 94.2–106%). Moreover, some authors report concentrations that have a high degree of uncertainty (*e.g.*, 0.289 µg L<sup>-1</sup>), and therefore are not acceptable to the analytical chemistry community.

### Abbreviations

AAS	Atomic absorption spectroscopy
ABS	Acetate buffer solution
BiFE	Bismuth film electrode
CCE	Carbon cloth electrode
CMC	Carboxymethyl cellulose
CNT	Carbon nanotube
COF	Covalent–organic framework
CPE	Carbon paste electrode
EDTA	2,2',2",2"'-(Ethane-1,2-diyldinitrilo)tetraacetic acid
GCE	Glassy carbon electrode
GO	Graphene oxide
IIP	Ion imprinted polymer
ICP MS	Inductively coupled plasma mass spectrometry
LOD	Limit of detection
LOQ	Limit of quantification
MOF	Metal–organic framework
MILs	Materials of Institute Lavoisier
MWCNTs	Multiwalled carbon nanotubes
NMs	Nanomaterials
NPs	Nanoparticles
PANI	Polyaniline
PBS	Phosphate buffer solution
PDA	Polydopamine
PEDOT	Poly(3,4-ethylenedioxythiophene)
PGE	Pencil graphite electrode
РРу	Polypyrrole
rGO	Reduced graphene oxide
SPE	Screen-printed electrode
SWASV	Square wave anodic stripping voltammetry
SWCNTs	Single walled carbon nanotubes
UiO	Universitetet i Oslo
ZIFs	Zeolitic imidazolate frameworks

### Conflicts of interest

There are no conflicts to declare.

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