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2D Materials-Based Platforms for Electroanalysis Applications

Georgina Alarcon-Angeles^a, Manuel Palomar-Pardavé^b, Arben Merkoçi^{c,d,*}

^a Universidad Autónoma Metropolitana-Xochimilco, Departamento de Sistemas Biológicos, C.P. 04960, D.F. México

^b Universidad Autónoma Metropolitana-Azcapotzalco, Departamento de Materiales, Área Ingeniería de Materiales, Av. San Pablo #180, Col. Reynosa-Tamaulipas, CDMX C.P. 02200, Mexico

* arben.merkoci@icn2.cat

^c Catalan Institute of Nanoscience and Nanotechnology (ICN2), CSIC and BIST, Campus UAB, Bellaterra, 08193 Barcelona, Spain

^d ICREA – Catalan Institution for Research and Advanced Studies, Barcelona, 08010, Spain

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Abstract

A new class of nanomaterials called “2D materials” (2DMs) is attracting recently the electrochemical sensing field due to the unique physicochemical properties associated to their chemical structure, formed by ultra-thin layers. In this review, we summarize the recent advances in the electroanalysis area using 2DMs giving first a brief overview on the structure, synthesis and properties of these materials followed by the analysis of their advantages while used in the development of electrochemical sensors.

Keywords: 2Dmaterials, Bio-sensors, Electroanalysis

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1. Introduction

During the last two decades the emerging of new nanomaterials (NMs) such as inorganic and organic nanoparticles, carbon nanotubes among others have been intensively explored in electroanalysis field. The large surface area and excellent electronic/mechanical/catalytic properties of NMs have remarkably improved electroanalysis research and development with interest for several applications [1-6].

Within various NMs a new class called “2D materials” (2DMs) are attracting, recently, the electrochemical sensing field due to the unique physicochemical properties associated to their chemical structure, formed by ultra-thin layers. 2DMs represent two-dimensional nanomaterials formed by atomic sheets (see Figure 1) with extraordinary properties; there are 150 exotic layered materials that can be easily split into a subnanometer-thick material [7]. 2DMs can also be classified as graphene, graphene oxide, hexagonal boron nitride (HBN), metal and non-metal chalcogenides, oxides, hydroxides and halides, covalent organic frameworks, silicates, and perovskites.

2D transition metal dichalcogenide (TMDs) include molybdenum disulphide (MoS₂), molybdenum diselenide (MoSe₂), tungsten disulphide (WS₂), and tungsten

diselenide (WSe₂), borophene (2D boron), black phosphorus, silicene (2D silicon), germanene (2D germanium), and MXenes (2D carbides/nitrides). Depending on their chemical compositions and structural configurations 2DMs can be categorized as metallic, semi-metallic, semiconducting, insulating, or superconducting, being the last property especially attractive for the development of electrochemical sensors [7].

In this review, we summarize the recent advances in the electroanalysis area using 2DMs giving first a brief overview on the structure, synthesis and properties of these materials followed by the analysis of their advantages while used in the development of electrochemical sensors.

2. Structure of 2DMs

In general, 2DMs are crystalline solids with a high side-size ratio (1-1000 μm) and thickness (< 1nm). These form strong intra-layer interaction of covalent bonds and Van der Waals interactions inter-layers. Strong covalent bonds give stability to the 2D planes, and the crystal structure of Van der Waals is sufficient to form heterostructures.

One of the most reported 2DMs is graphene. Its structure consists in a monolayer of carbon atoms with sp²

hybridization, whose geometry is like a honeycomb (Figure 1a), carbon nanotubes or fullerenes, can be obtained from the graphene monolayers. The structure of graphene is analogous to benzene and polycyclic rings, so the graphene chemistry is similar to these compounds.

On the other hand, the structure of graphene oxide (GO) is assumed to be a graphene sheet bonded to oxygen in the form of carboxyl, hydroxyl or epoxy groups. It has been proven that the GO sheets have an average roughness of 0.6 nm and the structure is predominantly amorphous due to distortions from the high fraction of sp^3 C–O bonds [8].

Black phosphorus, also known as phosphorene, belongs to the family of the two dimensions materials. The honeycomb-shaped geometric structure of the material is due to the formation of covalent bonds with three neighbouring phosphorus atoms (Figure 1c).

The structure of the molybdenum sulphide (MoS_2) is the most used of the TDMs. In its structure each plane is made of molybdenum atoms sandwiched between sulphur atoms (Figure 1d). MoS_2 layers structures have unique electronic, optical, mechanical and chemical characteristics like graphene. MoS_2 has a large surface area, which makes it advantageous for biosensing applications

A peculiarity of the 2DMs is that they can form heterostructures with layers of varied materials and with a thickness of one or two atoms, and thus synergistically improve their physicochemical properties.

(Figure 1. Near here)

Fig 1. Crystal structures (top view and side view) of (a) graphene, (b) silicene, and (c) phosphorene d) Chalcogenides e) boron nitride. a-c) From reference [9] with permission from RSC.

3- Synthesis

The most relevant methodologies for 2DMs production is wet chemical synthesis, the chemical vapor deposition (CVD), mechanical exfoliation, mechanical cleavage, epitaxial growth, and electrochemical exfoliation, liquid phase ultrasonic exfoliation [10-16]. For each preparation technology the control of the surface size (lateral size, thickness) and composition is quite important.

Liquid phase reactions, such as chemical exfoliation and hydrothermal or solvothermal reactions, have high efficiency, but the size of the sheets are usually small, and the quality of the product is relatively low and difficult to control compared with mechanical exfoliations.

The simplest form of CVD to grow 2DMs is the co-evaporation of metal oxides and chalcogen precursors that lead to vapor phase reaction followed by the formation of a stable 2D material over a suitable substrate. The growth mechanism of the CVD method differs in each synthesis process as the materials forming process also depends on (a)

properties of the substrate, (b) temperature, and (c) atomic gas flux [7]. The efficiency is much higher than that for mechanical exfoliation, and the quality control is better than the liquid phase routes. Therefore, CVD has been recognized as a reliable route for preparing high quality two-dimensional (2D) materials. Recently a review about the synthesis using this technique has been reported by Chang and Mannix [11-12].

Several strategies, including electrochemical techniques, have been investigated for improving the innate features of 2DMs. The Electrochemical Atomic Layer Deposition (E-ALD) is one of techniques that produces thin films of semiconductor. Exploiting Surface Limited Reactions (SLR), it enables the deposition of highly ordered an ultra-thin film from diluted aqueous solutions at room temperature.

Underpotential deposition (UPD) is a type of electrochemical SLR where an atomic layer of a metal is deposited onto a different nature substrate at potential values more positive than the equilibrium potential, so that the resulting deposit is generally limited to an atomic adlayer.

UPD has been used for electrodeposition of metals, such as Cu on Au(111) [1] and onto Au nanoparticles [17]. In order to study the initial stages of the formation and growth of metal it is important to understand the influence of the substrate's structure on the mechanism and kinetics of the monolayer formation, which entail the formation of a new 2D phase growing on the surface of the substrate [17].

To define and characterize the mechanism and kinetics involved during the deposition process in quantitative way traditional electrochemical techniques such as potentiostatic current transient measurements can be used [17,18]. It offers a significant advantage since the obtained data is easily interpreted with the aid of several theoretical models developed for this purpose.

On the other hand, graphene, graphene oxide [15] and Black phosphorus (BP) can be synthesized by mechanical exfoliation or by phase-liquid exfoliation, the thickness of the monolayers can reach sizes of 0.9 nm [19].

4. Physicochemical properties

Furthermore, 2DMs have also attracted attention because of their high degree of anisotropy and chemical functionality, which allows their application in different fields. Their biocompatibility and excellent electrical properties are very interesting too in sensors field.

2DMs, are strong, light, transparent, and flexible. They are excellent conductors of heat and electricity. They have a high conductivity, in addition to their excellent attributes in terms of electrochemical changes at the electrode interface. The modification of electrodes with 2DMs can produce a synergetic effect that improves the electrocatalytic activity,

conductivity, selectivity and biocompatibility while used in sensors.

These materials are uniquely suited for fundamental and applied electrochemical studies. Due to the extreme thinness their surface area is extremely large and the potential drop at the solid/liquid interface is “felt throughout the 2D interior” as well.

(Figure 2. Near here)

Fig 2. Techniques used for the synthesis of 2DMs. a) Mechanical exfoliation, b) physical vapour deposition c) chemical vapour deposition, From reference [12] with permission from Macmillan d) electrochemical exfoliation From reference [13] with permission from Macmillan, and e) phase-liquid exfoliation, From reference [10] with permission from Wiley-VCH, f) schematic production of chemically modified graphene from graphite. From reference [15] with permission from RSC, h) Schematic of the preparation of MoS₂-graphene hybrid nanosheets From reference [16] with permission from Wiley-VCH

Graphene oxide draws the attention due to its solubility in water, and other solvents allowing it to be uniformly deposited onto a wide range of substrates in the form of thin films or networks which makes it potentially useful for electrodes and other devices.

Black phosphorus (BP) presents fascinating properties as anisotropy, electric conductivity, supercapacity, and a wide cathodic window potential, however it is limited as a result of its low range of anodic potential, because it oxidizes at 0.6 V [20]. One advantage is its fast transfer heterogeneous and excellent absorption in the UV, which distinguishes it from other 2DMs. It has great stability at room temperature and low toxicity. Another property of BP is its ability to increase the band gap in 2eV. This depends on the number of layers and the size of particle, so the resulting electronic properties would depend on these features.

5. Applications

Over the years the challenge in the chemical and biological analysis is to develop new low-cost devices, which can be used for control and monitoring of the quality of the environment, food and health. The possibility that these devices may enable fast and reliable measurements of different compounds (ex. biomarkers or toxic compounds) could make the difference between life or death.

Nanotechnology offers new horizons for the application of nanomaterials in electrochemical sensing, which as is well known, have many advantages including low cost, high selectivity, and sensitivity. Nano-sized materials, given their high surface area, which in turn brings great advantages in terms of electronic, mechanical, and catalytic properties are considered of special interest in electrochemical sensors.

Electroanalysis in general and particularly electrochemical sensors and biosensors are showing to be an interesting alternative to conventional analytical techniques. These devices combine easy operation, low cost, rapid response, accuracy, sensitivity, and selectivity with the miniaturization and portability of the instruments, which are convenient for the real-time analysis of various compounds.

5.1. Sensors based on 2D nanomaterials

To date electrochemical sensors have been developed for a wide variety of compounds, among which are: glucose, neurotransmitters, environmental pollutants, such as heavy metals and pesticides among others.

The challenge for the construction of these devices lies in their stability, selectivity, and average life time, so over the last decade the usage of new materials has given a great possibility to improve and propose new electrochemical sensors and biosensors.

The inclusion of 2DMs as platform for electrochemical sensors have provided many benefits including easy functionalization and connection with receptors to improve selectivity. Table 1 presents a summary of various examples showing 2DMs application in electroanalysis.

Graphene (GR) is the most popular nanomaterial used in biosensors. Its application in biosensors is bringing advantages in terms of simplicity in addition to offering of novel detection opportunities in either optical or electrical-based techniques [21,22].

Usually the modification of the electrode's surface with GR can be achieved by covalent or non-covalent interactions; however, bonds of functional groups can destroy GR's conjugation and affect the material properties [23].

Polymers like chitosan have been used in combination with GR due to their high permeability and adhesion strength that contributes to the sensor selectivity. Graphene has been employed as platform for glucose, dopamine, H₂O₂, and heavy metals detection between others.

Glucose detection based on modified non-enzymatic electrode has been possible by using graphene [24,26]. This strategy can detect 2.5 nM of glucose when the composite is based on nickel nanoparticles incorporated within graphene nanosheets/graphene nanoribbons showing high electrochemical activity towards oxidation of glucose, good reproducibility and stability [26]. The glucose detection using graphene also is connected with a smartphone device. The system is composed by modified electrodes with graphene oxide, portable electrochemical detector and a smartphone that was tested for glucose measurements in blood serum samples showing reliable performance for detection of glucose in real time [27], Figure 3a.

(Figure 3. Near here)

Fig 3. Graphene strategies for detection of: a) Glucose via non-enzymatic mode. From reference [27] with permission from Elsevier, b) Cd (II), Pb(II), Fe (II) detection at glassy carbon electrode (GCE) modified with calixarene and reduced graphene oxide (CA/RGO) From reference [31] with permission from Elsevier.

Due to good adsorption of contaminants such as heavy metals onto graphene applications as ultrasensitive sensor for heavy metals including Cd(II), Pb(II), Zn(II), Hg(II) [28,30] are reported. (see Table 1.) Recently reduced graphene oxide (RGO) has been utilized for simultaneous voltammetric

detection of metal ions, Fe(II), Cd(II) and Pb(II) [31], reaching detection limits of 2 pM. This device was used for metal detection in pharmaceutical formulation with the

advantage that it is related to eco-friendly and highly efficient nanomaterial (Figure 3b).

Heavy metals preconcentration capability of graphene followed by electrochemical stripping analysis of heavy metals (ex. lead) in a lab-on-a-chip platforms also is demonstrated [32], Figure 4.

(Figure 4. Near here)

Fig 4. Lab-on-a-chip system for heavy metals preconcentration and response of preconcentration and detection of the Pb(II) From reference [32] with permission from ACS.

Table 1. Electroanalysis based on 2DMs

2D material	Analite	Platform	LOD	Detection Technique	Sample matrix	REF.
Graphene	Glucose	NiONFs/GO/GCE	0.77 μ M	Amperometry	Human blood serum	[24]
		NiNPs/ATP/RGO-GCE	0.37 μ M	Amperometry	Human serum	[25]
		GS/GNR/Ni/GCE	2.5 nM	Amperometry	---	[26]
		rGO/SPE	0.026mM	CV	----	[27]
	Dopamine 5-hydroxytryptamine	GR/p-AHNSA/SPCs	2 nM 3 nM	SWV	Pharmacological formulations, human urine and blood human serum	[37]
	Dopamine	2D g-C ₃ N ₄ /CuO/GCE	0.1 nM	SWV	----	[38]
	Uric acid	Fe ₃ O ₄ NPS/GNS/GCE	0.41 μ M	DPV	Brain tissue, urine, whole blood	[39]
	Cd(II) Pb(II)	TRGO/Au	1 μ g/L 0.4 μ g/L	SWASV	Drinking water	[40]
	As (III)	NH ₂ /GO/Au	0.162 ppb	SWASV	Groundwater	[41]
	H ₂ O ₂	rGO/PPyNT	100 pM	Amperometry	Synthetic solution of biological fluids	[42]
RGO-MWCNT-Pt/Mb		6 pM	Amperometry	----	[43]	
MoS ₂ /graphene		0.12 μ M	Amperometry	Water	[36]	
Black phosphorus	Dopamine Ascorbic Acid	BP		CV	---	[20]
	Pb(II) Cd (II) AsO ₄ ²⁻ Hg(II)	BP/metal electrode	1 ppb 3 ppb 10 ppb 1 ppb	Amperometry	----	[34]
	H ₂ O ₂	BP/GCE	1x10 ⁻⁷ M	Impedance	-----	[33]
	TDMs	H ₂ O ₂	BP//MoS ₂ -rGO/GCE	0.1 μ M 0.3 μ M	Amperometry	River water Tap water
MnO ₂ nanosheets/GCE			5 nM	Amperometry	Living cells	[44]
Glucose		NiSe ₂ -NS/GCE	23 nM	Amperometry	-----	[35]
Dopamine Uric acid		MoS ₂ /rGO/GCE	0.06 μ M 0.46 μ M	DPV	Serum samples	[45]
Dopamine		AuNPs/MoS ₂	80 nM	DPV	---	[46]
Dopamine Ascorbic acid Uric acid		MoS ₂ -rGO/GCE	0.12 μ M 0.09 μ M 0.10 μ M	DPV	---	[47]

	Clenbuterol	MoS ₂ -Au-PEI	1.92 ng/mL	DPV	Pork	[48]
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NA/NiONF-GO/GCE: Nafion/NiO nanofibers/ graphene oxide/ glassy carbon electrode

GS/GNR/Ni, Graphene sheet/graphene nanoribbon/nickel nanoparticles;

GR/p-AHNSA/SPCs: Graphene/4-amino-3-hydroxy-1-naphthalenesulfonic acid/Screen-printed carbon sensors

2D g-C₃N₄/CuO:Graphitic carbon nitride

Fe₃O₄NPs/GNS ferroferric oxide nanoparticles/ graphene oxide nanosheet

TRGO/Au : reduced graphene oxide/ gold electrode

NH₂-GO/Au amino-functionalized graphene oxide/ gold microelectrode

rGO/PpyNT: reduced graphene oxide/polypyrrole-nanotubes

BP: Blackphosphorus

NiSe₂-NS/GCE: NiSe₂ nanosheets/glassy carbon electrode

MoS₂/rGO /GCE: MoS₂/reduced graphene oxide/ glassy carbon electrode

MoS₂-Au-PEI/GCE: MoS₂-Au-polyethylenimine/ glassy carbon electrode

One of the newest 2D material used for sensing is black phosphorus (BP). Recently it has been used for non-enzymatic detection of hydrogen peroxide by electrochemical impedance. The response of the BP-modified electrode was considerably better than that of other electrodes. However, one drawback is the degradation of modifying layer **due** to oxidation of BP by H₂O₂ [33].

Some strategies have been developed for the detection of heavy metals using a mechanically exfoliated sheet of black phosphorus. In order to develop a sensitive sensor for heavy metals such as As (III), Pb (II), Cd(II), or Hg. This sensor was able to detect heavy metals down to ppb levels [34].

Because of high electrocatalytic activity, TDM has received great attention. Particularly materials like NiSe₂ which allow a fast electron transfer ensure effective response and very low limit of detection [35]. MoS₂-graphene composite exhibited enhanced electron transfer, kinetics and excellent electro-reduction performance towards H₂O₂ in alkaline media [36].

5.2. Biosensors based on 2D nanomaterials applications

Advances in the development of these devices highlight the inclusion of biological recognition material, such as enzymes, antibodies, and DNA probes. The 2DMs are also highlighted, for their good biocompatibility, the high surface area that increases the immobilization of a greater number of biomolecules per unit of area improving the efficiency of the biosensor.

One of the major challenges for the construction of a biosensor is the stability of the biomaterial for which a robust, stable, and efficient biocatalytic support platform is **necessary**. In the case of enzymatic biosensors, the key to a good device is the immobilization of the enzyme onto the electrode surface. It has been reported that with the usage of graphene the denaturation of the enzyme is avoided, and the thermodynamic stability is improved generating in this way an excellent biocatalysis [49-51]. For this reason graphene has been used as immobilization platform of various enzymes (ex. glucose oxidase, tyrosinase, cholesterol

oxidase, catalase, horseradish peroxidase, acetylcholinesterase) with interest for the detection of several analytes (respectively glucose, phenol, cholesterol, **alcohol**, hydrogen peroxide, pesticide). Graphene has shown good biocompatibility and high electrocatalytic activity, in addition to high sensitivity, and very low detection limits (see Table 2).

The 2D carbon nanomaterials, such as graphene oxide and carbon nanosheets have been widely studied to achieve the direct electrons transfer between enzymes and defects/edge sites while oxygen of functional groups could play an important role in the adsorption and orientation of the enzyme. As claimed thin films can provide an affine microenvironment for enzymes and thus improve the direct electrons transfer between enzymes and electrodes [50]. Current trends in biosensor is to develop electrodes of third generation based on 2DMs, Figure. 5a-g.

On the other hand, affinity-based assays, which use specific antibodies (Figure 5h), DNA or RNA for a target protein or small molecule offer a facile and rapid approach to biosensing with potential for high sensitivity and selectivity. An interesting application has been coupling of graphene with aptamers. Given the interest in building flexible sensors based on graphene, the use of aptamers as receptors for different targets (either small molecules or biological, organic or inorganic macromolecules) has been an advantageous approach [52,53].

(Figure 5. Near here)

Fig. 5. MoS₂/Au NPs/GOx biosensor a-d) Characterization, f) schematic representation of modified electrode, g) electrochemical response. From reference [51] with permission from Elsevier h-i) Schematic of the MoS₂ biosensor device j) electrochemical response. For biosensing, the MoS₂ is functionalized with receptors for specifically capturing target biomolecules From reference [52] with permission from RSC.

Another interesting application of graphene has been its use for removal of contaminants in lab-on-a-chip with

electrochemical detection of the same compound (removal of polybrominated diphenyl ethers, PBDEs) [53], Figure 6.

(Figure 6. Near here)

Fig. 6. Components of PDMS-reduced graphene oxide (rGO) chip, for the simultaneous detection and removal of polybrominated diphenyl ethers. From [53] with permission from Springer.

Table 2. Biosensors based on 2DMs

2D material	Analyte	Electrode	Bioreceptor	LOD	Detection Technique	Sample matrix	REF.
Graphene	Polybrominated diphenyl ethers (PBDEs)	rGO/SPE	Anti-PBDE	0.019 ppb	SWASV	--	[53]
	Glucose	GOD-Gr/PANI/AuNPs	GOD	0.6 μM	Amperometry	Human serum	[54]
	Parathyroid hormone	ALP-PTH-MG	MAb targeting PTH	--	CV	Human serum	[55]
	Cholesterol	CAT/ChOx/GR-IL/GCE	ChOx	0.05 μM	Amperometry	Human serum	[56]
	Dichorvos	Er-GRO-Nafion/ACHe	AChE	2.0 ng mL ⁻¹	Amperometry		[57]
	Paraoxon	RGON film	AChE	1.37 x10 ⁻⁷ M	Amperometry		[58]
	Methyl Parathion Chlorpyrifos Carbofuran	NiO NPs-CGR-NF/AChE	Acetylcholinesterase	5x10 ⁻¹⁴ M 5x10 ⁻¹³ M 5x10 ⁻¹³ M	Amperometric	cabbage apple, tap water and lake water	[59]
	AFP	Au-PDA-THi-GO	AFP-Ab	0.03 ngL ⁻¹	DPV	Human serum	[60]
	Prostate Cancer Biomarker	R-GO	PSA mAb	1.1 fM	Conductivity	Human serum	[61]
	H ₂ O ₂	HRP-MoS ₂ -Gr RGO-MWCNT-Pt/Mb	HRP	0.049 μM 6 pM	Amperometry Amperometry	--- ---	[62] [63]
Catechol	SPE/oGO/Tyr	Tyrosinase	0.1 nM	Amperometry	---	[64]	
TDMs	Glucose	MoS ₂ /Au NPs/GOx		0.042 μM	Amperometry		[51]
	Rabbit IgG	GCE/MoSe ₂	Anti rabbit IgG	1.23 ngmL ⁻¹	Amperometry	-----	[65]
	Fenothrothion	GC/WS/GA/AChE/BSA	AChE	2.86 nM	Amperometry	Apple juice	[66]
	H ₂ O ₂	AuMoS ₂	IgG-HRP	0.1 ng L ⁻¹	CV	---	[52]
	DNA	AuNPs/MoSe ₂ -Gr/GCE	DNA	4 fM	DPV	Human serum	[67]
Black phosphorus	Myoglobin-specific aptamer	BP	Aptamer-functionalized PLL-BP	0.524pgmL ⁻¹	CV	Human serum	[68]

GOD: Glucose oxidase

GO: Graphene oxide

AChE: acetylcholinesterase

Tyr: Tyrosinase

RGO/MECNT/Pt/Mb: reduced graphene oxide/multiwalled carbon nanotubes/myoglobin

ALP-PTH-MG: Parathyroid hormone, alkaline phosphatase, MoS₂-graphene; Mab /PTH: monoclonal antibody/ targeting

HRP-MoS₂-Gr: horseradish peroxidase

AFP: Alpha-fetoprotein

SWASV: Square wave anodic stripping voltammetry.,

DPV: Differential pulse voltammetry

SWV Square wave voltammetry

CV: Cyclic voltammetry

The interesting electrochemical properties of TDMs have been shown for example during modification of electrodes with MoS₂ nanosheet. Such modification was able to enhance electrocatalytic reactions, have a high faradic-to-capacitive current ratios as well as high current density. TDMs can be effectively tuned through different strategies such as reducing their dimensions, intercalation, heterostructuring, alloying and hybridising with secondary elements [51]

Frequently MoS₂ is used for hydrodesulfurization reaction as a catalyst, but the use of MoS₂ as an enzymatic biosensor favours the direct electron transfer. In order to improve the detection limit a sandwich immunoassay for H₂O₂ detection by immobilization of horseradish peroxidase conjugated

IgG via the electrostatic attraction has been proposed (Figure 5 h-i) [52].

The sensor exhibited high specificity and sensitivity imparted by the synergy of high affinity screened aptamers and the enhanced electrochemical properties of the nanocomposite. The sensor has a very low limit of detection of ~0.524 pg mL⁻¹ with a sensitivity of 36 $\mu\text{A pg}^{-1} \text{mL}^{-1}$ for Mb spiked in serum samples. This device can open up opportunities to realize better cardiac biomarker detection for a point-of-care diagnosis [54].

6. Conclusions and perspectives

Research and development in the field of 2DMs-based electroanalysis in general and biosensors particularly is growing significantly. The emergence of these new materials with interesting mechanical, electrical and optical properties are of special interest in order to create new cost efficient devices. Use of 2DMs have allowed to improve various parameters of electroanalysis devices and systems such as a) the enzyme catalytic properties and the reaction kinetics; b) enzyme immobilization achieving third generation enzymatic biosensors; c) Improved analytical performance parameters like sensitivity, affinity, detection and quantification limits.

Biosensors based on 2DMs in different configurations have demonstrated to be excellent alternatives for the determination of different compounds although a careful comparison with devices based on the use of other nanomaterials would better clarify such advantages.

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