# Application of 2D MoS2-Based Nanocomposites 

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The 2D molybdenum disulfide $\left(\mathrm{MoS}_{2}\right)$ nanosheets have unique and complementary properties to those of graphene, rendering them ideal electrode materials that could potentially lead to significant benefits in many electrochemical applications. These properties include tunable bandgaps, large surface areas, relatively high electron mobilities, and good optical and catalytic characteristics.

Keywords: Molybdenum disulfide ; Functionalization ; Metal and metal oxide nanostructures ; Electrochemical sensors ; Biosensor ; Detection ; Nanosheets

## 1. Introduction

Layered materials have been studied for several years. Each layered material, when thinned to its physical limit, reveals novel properties different from its bulk counterpart. Therefore, at the physical limit, these materials are referred to as twodimensional (2D) materials ${ }^{[1]}$. These 2D materials have received great attention due to the affluence of unusual physical characteristics that occur when charge and heat transports are confined to a plane [2]. Graphene is the most widely studied 2D material because of its exceptional properties. It is composed of a single layer of carbon atoms arranged in a 2 D honeycomb lattice ${ }^{[3]}$. It is a fundamental building block for a range of well-known carbon materials such as threedimensional (3D) graphite, one-dimensional (1D) carbon nanotubes, and zero-dimensional (OD) fullerene ${ }^{[3]}$. Since its isolation, it has inspired thoughtful research on other 2D materials. These materials are useful building blocks that can be restacked and incorporated into composites for a wide range of applications. Other than graphene, recent efforts have focused on other 2D materials (ranging from conductors to insulators). These materials include transition metal dichalcogenides (TMDs) as semiconductors involving a general chemical formula of $M X_{2}$, where $M$ is a transition metal atom ( $\mathrm{M}=\mathrm{Mo}, \mathrm{W}, \mathrm{Ti}, \mathrm{Zr}, \mathrm{Ta}, \mathrm{Nb}, \mathrm{Re}, \mathrm{Ni}$, or V ) and X is a chalcogenides atom ( $\mathrm{X}=\mathrm{S}, \mathrm{Se}, \mathrm{Te}$ ), as well as black phosphorus (BP), transition metal oxides, and hexagonal boron nitride (hBN) as an insulator [3][4][5]. TMDs are now the key focus of many researchers because of their unique properties. Exfoliated 2D TMDs have properties that are complementary to (but yet distinct from) those of graphene, and they also have the advantages of tunable bandgaps ranging from 1-2 eV [4][6][7]. One of the most widely explored transition metal dichalcogenides is molybdenum disulfide $\left(\mathrm{MoS}_{2}\right){ }^{[8]}[9][10]$. The electrical and optical properties of $\mathrm{MoS}_{2}$ are layer-dependent. For instance, bulk $\mathrm{MoS}_{2}$ is an n-type semiconductor with an indirect bandgap of about 1.3 eV , while a monolayer $\mathrm{MoS}_{2}$ has a direct bandgap of 1.8 eV [6][11]. Moreover, monolayer $\mathrm{MoS}_{2}$ exhibits carrier mobility of $200 \mathrm{~cm}^{2} \mathrm{~V}^{-1} \mathrm{~s}^{-1}$ at room temperature with a high on/off current ratio of $\sim 10^{8}\left[\underline{12]}\right.$. MoS $\mathrm{Mas}_{2}$ ha layered crystal structure formed by stacking covalently bound S-Mo-S monolayers through weak van der Waals interactions ${ }^{[13][14]}$. The van der Waals interlayer interaction between these monolayers allows for their separation leading to crystal exfoliation with approximately 0.65 nm of thickness $(6.5 \AA){ }^{[12][15]}$.

As a semiconducting analog of graphene, this material has recently been reported as promising in the application of sensors ${ }^{[13]}$, photodetectors ${ }^{[16]}$, transistors ${ }^{[12]}$, flexible electronics ${ }^{[17]}$, and fuel cells ${ }^{[18]}$. Although the use of $\mathrm{MoS}_{2}$ nanosheets offers many advantages, poor conductivity (due to the large bandgaps) considerably limits their practical applications in sensors. Moreover, it has been reported that pristine $\mathrm{MoS}_{2}$ nanosheets suffer from gradual degradation at ambient conditions, which are triggered by surface contamination and significant adsorption of oxygen in an air environment. This leads to instability and decreased electrical properties and sensing abilities [19][20]. Therefore, to obtain satisfactory results, pristine $\mathrm{MoS}_{2}$ nanosheets need to be operated in an inert atmosphere. To overcome these limitations and extend the potential application of $\mathrm{MoS}_{2}$ nanosheets, various approaches have been attempted, such as the substitution of the transition metal of $\mathrm{MoS}_{2}$ with other elements [21], molecular physisorption of organic materials [22], changing interfacial chemistry ${ }^{[23][24]}$, solution-based chemical doping ${ }^{[25]}$, and promoting structural phase transition (from semiconducting 2 H phase to metallic 1 T phase) ${ }^{[26][27] \text {. However, some of these methods are complicated and time- }}$ consuming; some result in structural defects, reduced mobility, and unstable composites [28]. Furthermore, researchers noted that TMDs have a habit of being inert to chemical functionalization. Chalcogen atoms in the basal plane of TMD nanosheets are saturated and, therefore, are not highly reactive, whereas the metal sites in TMDs are embedded beneath
the chalcogen layer, all but eliminating them from being useful for functionalization [29]. Despite this, it is necessary to functionalize $\mathrm{MoS}_{2}$ with chemical moieties, which can enable its interface with other nano- or micro-structures [15]. The synergetic effects resulting from $\mathrm{MoS}_{2}$ composites can produce enhanced properties or improved performances. The large surface-to-volume ratio of $2 \mathrm{D} \mathrm{MoS}_{2}$ nanosheets presents an opportunity for the effective surface functionalization of the material with metal and metal oxide nanostructures (NSs). Compared with previously described methods of functionalization, interfacing 2 D MoS 2 nanosheets with metal-based NSs is more simply carried out and offers an additional path for controlling the thermal, catalytic, magnetic, optical, and electrical properties. Based on theoretical calculations, it has been reported that the effective adsorption of different metal atoms on 2 D MoS 2 is related to the number of $d$
-electrons, which can also effectively modulate the band structures of the material [30]. Most metal-based NSs have good stability and are resistant to environmental deterioration and oxidation, which make them ideal for the fabrication of nanocomposites. However, it is worth noting that the structures and properties of nanocomposites are dependent on the conditions of the composite synthesis and the control of the decorated location, morphology, and number density of the NSs ${ }^{[31]}$. Recently, metal and metal oxide nanocomposites have received a great deal of attention because of their excellent physical and chemical properties, which expanded their application in sensors ${ }^{[20][31]}$, electrocatalysis $\frac{[32][33]}{}$, and optoelectronics ${ }^{[34]}$. Thus, it can be expected that 2D MoS 2 nanosheets decorated with metal-based NSs could potentially extend their applications as novel nanomaterials in sensors. For instance, $2 \mathrm{D} \mathrm{MoS}_{2}$ functionalized with metal and metal oxide NSs , such as nickel ( Ni$)^{[35]}$, gold $(\mathrm{Au})^{[36]}$, and tricobalt tetraoxide $\left(\mathrm{Co}_{3} \mathrm{O}_{4}\right){ }^{[37]}$, to name a few, was reported to exhibit excellent catalytic sensing properties. The $\mathrm{MoS}_{2}-\mathrm{Ni}$ nanocomposite demonstrated good reproducibility and excellent sensitivity toward glucose detection. The reported results showed that small Ni nanoparticles (NPs) on the surface of the $\mathrm{MoS}_{2}$ nanosheet had more active sites, resulting in high electrocatalytic activity and a fast response time of less than 2 s . In addition, it was discovered that Ni NPs without $\mathrm{MoS}_{2}$ support appeared to aggregate, which was detrimental to the electrocatalytic activity of the sensor ${ }^{[35]}$. The $\mathrm{MoS}_{2}-\mathrm{Co}_{3} \mathrm{O}_{4}$ nanocomposites showed high sensitivity and fast response and recovery features for the detection of ammonia at room temperature. The sensing film was constructed on an interdigital electrode substrate using layer-by-layer self-assembly of $\mathrm{MoS}_{2}$ nanosheets and $\mathrm{Co}_{3} \mathrm{O}_{4}$ nanorods. The layer-by-layer self-assembly not only efficiently prevented agglomeration but also provided many more active catalytic sites on p-type $\mathrm{Co}_{3} \mathrm{O}_{4}$ nanorods toward ammonia. The results indicated that the fundamental sensing mechanisms of the $\mathrm{MoS}_{2}-\mathrm{Co}_{3} \mathrm{O}_{4}$ nanocomposite towards ammonia were attributed to the layered nanostructure, synergistic effects, and p -n heterojunction depletion layer formed at the interface of n-type $\mathrm{MoS}_{2}$ and p-type $\mathrm{Co}_{3} \mathrm{O}_{4}{ }^{[377]}$. These 2D-MoS $2 /$ metal-NSs composite sensor films showed significant improvements in sensitivity in comparison with the 2 D MoS 2 and metal/metal oxide nanostructure counterparts, suggesting that the large surface areas, high conductivity, and improved biocompatibility played significant roles in the resulting sensing outcome. Su et al. ${ }^{[10]}$, also reported that $\mathrm{MoS}_{2}$ stabilizes metallic NPs, such as platinum (Pt), Au, silver (Ag), and lead (Pd) to form hierarchical nanocomposites, and such 2D$\mathrm{MoS}_{2}$ /metal-NSs composites possess the essential properties of pure metal NPs and $\mathrm{MoS}_{2}$ nanosheets due to their synergistic effects, making the $2 \mathrm{D} \mathrm{MoS}_{2}$ and metal/metal oxide nanostructured composites exhibit excellent electrochemical properties for the fabrication of electrochemical sensors ${ }^{[13]}$.

## | 2. Synthesis of $\mathrm{MoS}_{2}$ Nanosheets

Different methods for synthesizing $2 \mathrm{D} \mathrm{MoS}{ }_{2}$ nanosheets have been proposed; these methods have also been used for tuning 2D $\mathrm{MoS}_{2}$ electronic band structures. So this creates an opportunity to find new methods that are appropriate for sensing applications. Lately, $\mathrm{MoS}_{2}$ nanosheets have been produced by either mechanical or chemical methods. In a standard mechanical exfoliation procedure, sufficient thin $\mathrm{MoS}_{2}$ crystals are first peeled off from their bulk crystals, layer-by-layer, using adhesive Scotch tape and exerting normal force. By repeating this process multiple times, the bulk $\mathrm{MoS}_{2}$ becomes thinner until it is reduced to a single sheet of $\mathrm{MoS}_{2}$. The cleaved thin crystals are detached with devices, such as plastic tweezers, and are adsorbed onto the target substrate ${ }^{[38]}$. While this method can yield pristine and high-quality 2D $\mathrm{MoS}_{2}$ nanosheets, poor scalability limits its practical application in general.

Chemical methods include:

1. lithium (Li) intercalation exfoliation, which involves an initial 48 h intercalation of Li ions between bulk $\mathrm{MoS}_{2}$ in an inert gas atmosphere followed by a spontaneous ultrasonic-assisted exfoliation of the Li-intercalated $\mathrm{MoS}_{2}$, which occurs through water reaction [39][40]. During the process, bulk $\mathrm{MoS}_{2}$ is treated with an $n$-butyllithium solution to produce the intercalation product $\mathrm{Li}_{\mathrm{x}} \mathrm{MoS}_{2}$. The reaction process is caused by the electron transfer from n-butyllithium to $\mathrm{MoS}_{2}$ layers, which absorbs $\mathrm{Li}^{+}$between the layers to balance the charge. When the intercalate, $\mathrm{Li}_{\mathrm{x}} \mathrm{MoS}_{2}$, comes into contact with water, it reacts with the intercalated lithium, creating LiOH and $\mathrm{H}_{2}$ gas. As a result, negatively-charged nanosheets repel each other, increasing interlayer distances and weakening van der Waals forces, resulting in a colloidal
dispersion of (nearly) entire single layers of $\mathrm{MoS}_{2}$ [41]. The lithium intercalation reaction converts $\mathrm{MoS}_{2}$ from the semiconductive 2 H phase hexagonal structure to the metallic trigonal 1 T phase ${ }^{[42]}$, although the 1 T phase $\mathrm{MoS}_{2}$ is not suitable for optoelectronic devices, it is quite desirable for electrochemical catalysis.
2. Liquid-phase exfoliation involves exfoliation by sonication [43][44] or exposure of bulk $\mathrm{MoS}_{2}$ to high shear rates using either a rotor-stator high shear mixer or a basic kitchen blender [45][46]. The bulk $\mathrm{MoS}_{2}$ is exfoliated in the presence of stabilizing liquids, such as suitable solvents [43], surfactant solutions ${ }^{[46]}$, or polymers ${ }^{[47]}$. Sonication-assisted exfoliation is triggered by hydrodynamic shear forces associated with cavitation, which is the formation, growth, and collapse of voids or bubbles in liquids due to pressure changes [44]. Following exfoliation, the inter-sheet attractive forces and interfacial tension between the sheets and the surrounding liquid are reduced, resulting in the dispersion of a single layer [44]. When using a high shear mixer or blender, exfoliation occurs as a result of revolving rotor-stator blades producing extremely high shear rates [45][46]. The shear rate generates full turbulence, resulting in viscous shear forces. The presence of dominant viscous shear forces increases collision and cavitation between bulk materials, leading to exfoliation ${ }^{[45][46]}$. During this process, the sheets are coated with an appropriate surfactant/solvent and stabilized by repulsive inter-sheet interactions ${ }^{[46]}$. This approach results in defect-free single or multilayered nanosheets that are stabilized against aggregation by liquid interaction. The liquid-phase exfoliation method, due to its simplicity and scalability potential, has an advantage over the lithium exfoliation method; it is also not an air-sensitive process and it does not require chemical reactions; thus, it provides high crystallinity for synthesized $\mathrm{MoS}_{2}$ nanosheets ${ }^{[48]}$.
3. Hydrothermal/solvothermal synthesis is essentially where the chemical reaction takes place in a closed system (autoclave) in which the solvent temperature is raised to its critical point ( $>200^{\circ} \mathrm{C}$ ) by heating concurrently with autogenous pressure [49]. This approach involves direct crystallization from solutions, which often include crystal nucleation and subsequent growth [49]. The sample is rapidly precipitated from the reaction solution during the synthesis process, allowing for controlled homogeneity, as well as control over aging, particle size, and morphology [50]. The solvothermal synthesis is similar to the hydrothermal synthesis, except that in the synthetic method organic solvents are used instead of water ${ }^{[51]}$. $\mathrm{MoS}_{2}$ nanosheets with good crystalline structure and morphology are produced by optimization of hydrothermal temperature and reaction time [50].
4. Chemical vapor deposition (CVD) has been used to synthesize high-quality graphene, and it was recently adopted for the synthesis of $\mathrm{MoS}_{2}$ nanosheets. In a standard CVD procedure, molybdenum trioxide $\left(\mathrm{MoO}_{3}\right)$ and sulfur powders are common precursors used for the deposition of $\mathrm{MoS}_{2}$ films on a silicon/silicon dioxide ( $\mathrm{Si}_{2} \mathrm{SiO}_{2}$ ) substrate; upon heating, $\mathrm{MoO}_{3}$ reacts with sulfur vapor in the gas phase at a high temperature ( $>650{ }^{\circ} \mathrm{C}$ ) to give $\mathrm{MoS}_{2}$ layers in a reducing atmosphere under ambient pressure ${ }^{[52]}$. The CVD process is capable of producing nanosheets of good quality with scalable size, controllable thickness, and excellent electronic properties. However, it is more difficult to obtain crystalline 2D $\mathrm{MoS}_{2}$ nanosheets with a controlled number of layers by CVD as compared to graphene, because the structure, thickness, and crystallinity of graphene could be well controlled by an effective catalyst design but there is no catalyst involved in the growth of $2 \mathrm{D} \mathrm{MoS}_{2}$ nanosheets [52][53].

The important thing to realize is that due to the nature of the exfoliation method, many imperfections, such as surface defects, may occur. The usual defect positions that occur are sulfur vacancies on the surface or edges [54][55]; due to these defects, $2 \mathrm{D} \mathrm{MoS}_{2}$ nanosheets have low charge carrier mobility and density [55]. These defects were recognized as possible synthetic targets for the functionalization and modification of $\mathrm{MoS}_{2}$ surfaces for various additional applications [54]. The functionalization of $\mathrm{MoS}_{2}$ is important to adjust or add the required properties to the material, which can be utilized in the fabrication of sensors and catalysts. This is an advantage given that 2 D MoS 2 offers abundant exposed edges that are known as the root of the catalytic activity ${ }^{[56][57]}$.

## 3. Functionalization of $\mathrm{MoS}_{2}$ Nanosheets with Metal and Metal Oxide Nanostructures

Metal and metal oxide NSs are known for their exceptional electrical and catalytic properties; hence, they are the most widely used nanomaterials. The application of these nanomaterials to medicine ${ }^{[58]}$, energy storage, catalysts, sensors [59] [60][61], and electronics ${ }^{[62]}$ has led research into the development of synthetic pathways toward the formation of nanocomposites. The integration of these NSs on $\mathrm{MoS}_{2}$ can effectively exploit the distinct qualities of both materials, which have already attracted considerable attention in the sensing field. The metal-based NSs can be distributed and significantly improved by the effective support matrix of $\mathrm{MoS}_{2}$ nanosheets, where the synergistic effects of metal-based NSs and $\mathrm{MoS}_{2}$ nanosheets can lead to greater catalytic efficiency and conductivity than plain $\mathrm{MoS}_{2}$. Several factors play significant roles in the properties of metal and metal oxide NSs, such as crystal structure, size, shape, morphology, and surface chemistry [63]. Thus, structuring methods have been applied to obtain different sizes and shapes of NSs with promising properties, such as high sensitivity, faster electron transfer kinetics, low background currents, high surface areas, and high current densities [64]. Metal and metal oxide NSs have been used to modify electrodes for use as
electrocatalysts in sensors; hence, they play significant roles in diagnostic devices. The $2 \mathrm{D}-\mathrm{MoS}_{2} /$ metal-NS composites utilize the optimum availability of the nanoscale surface area for electron transfer, and also enable mass transport of the reactants to the electroactive focal point on the electrode surface, resulting in a significantly improved electrochemical response [65][66].

The decoration of $\mathrm{MoS}_{2}$ nanosheets with metal-based NSs is commonly carried out in two different ways: in situ or ex situ functionalization. These functionalization methods can be achieved by post immobilization of NSs on $\mathrm{MoS}_{2}{ }^{[65]}$, hydro/solvothermal reaction ${ }^{[66]}$, electrodeposition ${ }^{[13]}$, or the chemical reduction method ${ }^{[35]}$. In these functionalization methods, the growth control of NSs on $\mathrm{MoS}_{2}$ nanosheets is a prerequisite for tuning the shape, size, and morphology. This growth control however involves precise process monitoring of the experimental parameters, such as the concentration of the reactant, time and temperature of the reaction, pH solution, the surfactant used, and the type of metal salts [67][68]. For instance, in a chemical synthesis process, the size and shape of the NPs can be effectively controlled by the concentration of metal salts, reaction conditions, or the use of different surfactants. Moreover, through an electrochemical synthesis, the concentration of surfactants, growth temperature, and current density can be optimized to control the size and shape of the NPs ${ }^{[68]}$. Concerning the subject, a study was conducted where thionine $\left(\mathrm{C}_{12} \mathrm{H}_{10} \mathrm{~N}_{3} \mathrm{~S}^{+}\right)$$\mathrm{MoS}_{2}$ was functionalized with AuNPs by a hydrothermal reaction for electrochemical immunosensing ${ }^{[69]}$. $\mathrm{C}_{12} \mathrm{H}_{10} \mathrm{~N}_{3} \mathrm{~S}^{+}$was used as a reducing agent and a surfactant to tune the resulting AuNPs structures on the surface of the $\mathrm{MoS}_{2}$ nanosheets. The study demonstrated that various well-defined shapes of AuNPs were produced by adjusting the concentration of $\mathrm{C}_{12} \mathrm{H}_{10} \mathrm{~N}_{3} \mathrm{~S}^{+}$while the concentration of metal salt $\left(\mathrm{HAuCl}_{4}\right)$ remained constant. It was discovered that with an increasing concentration of $\mathrm{C}_{12} \mathrm{H}_{10} \mathrm{~N}_{3} \mathrm{~S}^{+}$, the size of the NPs increased and the shape changed from spherical, triangle, and cloverlike to flower-like shape. A significant finding was that $\mathrm{C}_{12} \mathrm{H}_{10} \mathrm{~N}_{3} \mathrm{~S}^{+}$could not facilitate the growth of AuNPs in the absence of $\mathrm{MoS}_{2}$ nanosheets. This indicated that $\mathrm{C}_{12} \mathrm{H}_{10} \mathrm{~N}_{3} \mathrm{~S}^{+}$and $\mathrm{MoS}_{2}$ had a synergistic effect on the formation of AuNPs and the growth of AuNPs was also promoted by the effective support matrix of $\mathrm{MoS}_{2}$ nanosheets. The proposed mechanism was because $\mathrm{C}_{12} \mathrm{H}_{10} \mathrm{~N}_{3} \mathrm{~S}^{+}$is an electrochemical indicator, the growth of AuNPs on the surface of $\mathrm{MoS}_{2}$ nanosheets could be due to the redox reaction among $\mathrm{MoS}_{2}, \mathrm{C}_{12} \mathrm{H}_{10} \mathrm{~N}_{3} \mathrm{~S}^{+}$, and $\mathrm{AuCl}_{4}{ }^{-}$. $\mathrm{C}_{12} \mathrm{H}_{10} \mathrm{~N}_{3} \mathrm{~S}^{+}$was absorbed on the surface of $\mathrm{MoS}_{2}$ nanosheets through a $\pi-\pi$
interaction and an electrostatic interaction due to its planar aromatics structure. Therefore, $\mathrm{MoS}_{2} / \mathrm{AuCl}_{4}^{-}$and $\mathrm{C}_{12} \mathrm{H}_{10} \mathrm{~N}_{3} \mathrm{~S}^{+} / \mathrm{AuCl}_{4}^{-}$formed two redox pairs, enabling the spontaneous transfer of electrons from $\mathrm{MoS}_{2}$ and $\mathrm{C}_{12} \mathrm{H}_{10} \mathrm{~N}_{3} \mathrm{~S}^{+}$to Au ions, allowing the reduction of Au ions to AuNPs on the $\mathrm{MoS}_{2}$ nanosheets ${ }^{[69][70][71]}$.

Recently published reviews have reported on the synthesis and application of metal and metal oxide NSs [68][72]. The reviews included extensive information on the effects of structural morphology, size, and shape on the properties of NSs and their application in the field of electrochemical sensors. Interestingly, all studies clearly indicate that the shape and structural morphologies of NSs serve important roles in determining their resulting electrocatalytic properties, as the number of exposed catalytic active sites is specifically dependent on the shape of the NPs. That being said, it was also reported that a perfect shape-surface structure interaction is unrealistic and, even with a well-defined size and shape, its surface will be extremely complex. It consists of not only some ordered surface domains of different dimensions but also a defined number of defects, corners, edges, steps, and kink sites, all of which contribute to the resulting electrocatalytic activity [73].

Many methods for the preparation of size and shape-controlled metal-based NSs are now available in the literature. Various sizes, shapes, and structures can be synthesized, such as OD NSs, which include structures, such as NPs, quantum dots, and nanospheres, as well as 1D NSs with high aspect ratios, such as nanorods, nanocombs, nanofibers, nanotubes, nanoneedles, nanoribbons, and nanowires. The 2D NSs are known as nanoplates, nanosheets, and nanopellets, while nanocrystals, multi-nanolayers, nanoflowers, and snowflakes are characterized as 3D NSs [72][74]. A comprehensive effort has been made to control the shapes and sizes of metal-based NSs in order to guarantee their efficient performances in electrochemical sensors. The synthesis of various forms of NSs not only enhances physical and chemical properties but also improves the biocompatibility and bioefficacy for the immobilization of biomolecules [68][72][75]. Moreover, due to the many active sites provided by particular NSs, a wide linear range and low detection limits have been achieved, resulting in NSs being ideal materials for the fabrication of biosensor devices. Table 1 summarizes the various functionalization methods of $\mathrm{MoS}_{2}$ nanosheets with different metal and metal oxide NSs, their resulting structural morphologies (size and shape), and their application in electrochemical sensors. The commonly used technique for making 2D-MoS ${ }_{2}$ /metal-NSs composites is in situ chemical growth.

Table 1. Functionalization methods of $\mathrm{MoS}_{2}$ nanosheets with different metal and metal oxide NSs and their applications in electrochemical sensors.

|  | $\mathrm{MoS}_{2}$-NS <br> Composites | Method of Functionalization | Metal and <br> Metal Oxides <br> Structural <br> Morphology | Size <br> (Diameter) | Type of Sensor | Ref. |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| Ex situ functionalization | $\mathrm{MoS}_{2}-\mathrm{Au}$ | Post immobilization | Nanoparticles | 5 nm | Electrochemical biosensor | [65] |
|  | TTR-MoS 2 -Au | Post <br> immobilization | Nanocrystals | - | Photoelectrochemical immunosensor | [76] |
|  | $\mathrm{MoS}_{2}-\mathrm{PEI}-\mathrm{Au}$ | Post immobilization | Nanoparticles | 12 nm | Electrochemiluminescence immunosensor | [77] |
|  | $\mathrm{CuO}-\mathrm{MoS}_{2}$ | Post immobilization | Nanotubes | 20 nm | Electrochemical sensor | [78] |


|  | $\mathrm{MoS}_{2}$-NS <br> Composites | Method of Functionalization | Metal and <br> Metal Oxides <br> Structural <br> Morphology | Size <br> (Diameter) | Type of Sensor | Ref. |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| In situ functionalization | $\mathrm{MoS}_{2}-\mathrm{Au}$ | Electrodeposition | Nanoparticles | 10 m | Electrochemical aptasensor | [13] |
|  | $\mathrm{MoS}_{2^{-}}$ <br> MWCNT/Au | Electrodeposition | Nanoparticles | 3-5 nm | Electrochemical sensor | [79] |
|  | $\mathrm{MoS}_{2}-\mathrm{Au} / \mathrm{Pt}$ | Electrodeposition | Nanoparticles | 100 nm | Electrochemical biosensor | [80] |
|  | $\mathrm{Cu}-\mathrm{MoS}_{2}$ | Electrodeposition | Nanoflowers | - | Electrochemical biosensor | [81] |
|  | $\mathrm{ZnO}-\mathrm{MoS}_{2}$ | Electrodeposition | Nanosheets | 50 nm | Electrochemical sensor | [82] |
|  | $\mathrm{Ni}-\mathrm{MoS}_{2}-\mathrm{Naf}$ | Chemical reduction | Nanoparticles | 6 nm | Electrochemical sensor | [35] |
|  | $\mathrm{Au}-\mathrm{MoS}_{2}$ | Chemical reduction | Nanoparticles | 80 nm | Electrochemical sensor | [83] |
|  | $\mathrm{N} / \mathrm{F} / \mathrm{MoS}_{2}-\mathrm{Ag}$ | Chemical reduction | Nanoparticles | 3 nm | Electrochemical sensor | [84] |
|  | $\mathrm{Au}-\mathrm{Pd} / \mathrm{MoS}_{2}$ | Chemical reduction | Nanoparticles | - | Electrochemical sensor | [85] |
|  | $\mathrm{TiO}_{2}-\mathrm{MoS}_{2}-\mathrm{Au}$ | Chemical reduction | Nanoparticles | 5-10 nm | Photoelectrochemical aptasensor | [86] |
|  | $\mathrm{Pt}-\mathrm{MoS}_{2}$ | Chemical reduction | Nanoparticles | 2.5 nm | Electrochemical biosensor | [87] |
|  | PtNi-MoS ${ }_{2}$ | Chemical reduction | Nanoparticles | $\begin{gathered} 1.35-6.26 \\ \mathrm{~nm} \end{gathered}$ | Electrochemical sensor | [88] |
|  | $\mathrm{Cu}_{2} \mathrm{O}-\mathrm{MoS}_{2}$ | Chemical reduction | Nanoparticles | 6-18 nm | Electrochemical sensor | [89] |
|  | $\mathrm{PtW}-\mathrm{MoS}_{2}$ | Chemical reduction | Nanocubes | 10 nm | Electrochemical sensor | [90] |
|  | $\mathrm{Pd}-\mathrm{MoS}_{2}$ | Chemical reduction | Nanoparticles | - | Electrochemical aptasensor | [91] |
|  | PtPd-MoS 2 | Chemical reduction | Nanocubes | 50 nm | Electrochemical immunosensor | [92] |


|  |  | Metal and |  |  |  |
| :--- | :--- | :--- | :--- | :--- | :--- |
| $\mathrm{MoS}_{2}$-NS | Method of | Metal Oxides | Size | Type of Sensor | Ref. |
| Composites | Functionalization | Structural | (Diameter) |  |  |
|  |  | Morphology |  |  |  |


| $\mathrm{Pt}-\mathrm{MoS}_{2}$ | Chemical reduction | Nanoparticles | - | Electrochemical sensor | [93] |
| :---: | :---: | :---: | :---: | :---: | :---: |
| $\mathrm{Ag}-\mathrm{MoS}_{2}$ | Chemical reduction | Nanoparticles | 5 nm | Electrochemical sensor | [94] |
| $\mathrm{MoS}_{2}-\mathrm{Pt}$ | Chemical reduction | Clover-like nanoparticles | $\begin{gathered} 15.3-2 \\ \mathrm{~nm} \end{gathered}$ | Electrochemical sensor | [95] |
| $\mathrm{Au}-\mathrm{Pd}-\mathrm{Pt} / \mathrm{MoS}_{2}$ | Chemical reduction | Nanoflowers | $14-26 \mathrm{~nm}$ | Electrochemical sensor | [96] |
| PdNi-MoS 2 | Chemical reduction | Nanowires | 0.5-3 nm | Electrochemical sensor | [97] |
| $\mathrm{MoS}_{2}-\mathrm{Cu}_{2} \mathrm{O}-\mathrm{Au}$ | Hydrothermal reaction | Nanocrystals | 20-30 nm | Electrochemical immunosensor | [98] |
| $\mathrm{NiO}-\mathrm{MoS}_{2}$ | Hydrothermal reaction | Nanoparticles | 38-72 nm | Electrochemical sensor | [99] |
| $\mathrm{Fe}_{2} \mathrm{O}_{3}-\mathrm{MoS}_{2}$ | Hydrothermal reaction | Nanoflowers | - | Electrochemical sensor | [100] |
| $\mathrm{Fe}_{3} \mathrm{O}_{4}-\mathrm{MoS}_{2}$ | Hydrothermal reaction | Nanospheres | 20-30 nm | Electrochemical sensor | [101] |
| $\mathrm{MoS}_{2}-\mathrm{TiO}_{2}$ | Hydrothermal reaction | Nanorods | 20 nm | Photoelectrochemical biosensing | [102] |
| $\mathrm{Ag} / \mathrm{MoS}_{2} @ \mathrm{Fe}_{3} \mathrm{O}_{4}$ | Hydrothermal reaction | Nanospheres | 50 nm | Electrochemical immunosensor | [103] |
| $\mathrm{MoS}_{2}-\mathrm{Cu}_{2} \mathrm{O} / \mathrm{Pt}$ | Solvothermal reaction | Nanoparticles | $\begin{gathered} 15 \text { and } 3 \\ \mathrm{~nm} \end{gathered}$ | Electrochemical immunosensor | [66] |
| $\mathrm{Cu}-\mathrm{MoS}_{2}$ - Naf | Solvothermal reaction | Nanoparticles | $1-5 \mathrm{~nm}$ | Electrochemical sensor | [104] |
| $\mathrm{NiCo}_{2} \mathrm{O}_{4}-\mathrm{MoS}_{2}$ | Solvothermal reaction | Nanorods | - | Electrochemical sensor | [105] |

TTR; transthyretin, PEI; polyethylenimine, CuO; copper oxide, MWCNT; multiwalled carbon nanotubes, $\mathrm{Cu}_{2} \mathbf{O}$; cuprous oxide, $\mathbf{C u}$; copper, $\mathbf{Z n O}$; zinc oxide, Naf; Nafion, N/F; nitrogen fluorine, $\mathrm{TiO}_{2}$; titanium dioxide, PtW; platinum/tungsten, NiO ; nickel oxide, $\mathrm{Fe}_{2} \mathrm{O}_{3}, \mathrm{Fe}_{3} \mathrm{O}_{4}$; iron (II, III) oxide, $\mathrm{NiCo}_{2} \mathrm{O}_{4}$; nickel cobaltite.

## 4. Properties

The 2D nanomaterials are known to have special properties, which are distinct from their 3D bulk counterparts. The 2D $\mathrm{MoS}_{2}$ nanosheets with completely tunable properties can be synthesized successfully with various types of methods as discussed above. Due to changes in properties with a decrease in the number of layers, 2D MoS ${ }_{2}$ nanosheets possesses significantly distinctive electrical, optical, and chemical properties, which are known to play important roles in the fabrication of sensors. The exceptionally large surface areas of $2 \mathrm{D} \mathrm{MoS}_{2}$ nanosheets offer robust surface functionalization and provide new possibilities for functional devices based on 2D materials. $\mathrm{MoS}_{2}$ has been reported to simply form nanocomposites with metal and metal oxide NSs [65][78], carbon nanomaterials [106][107], conductive polymers [108][109][110], and other 2D nanomaterials [111][112], which could be used as catalytic materials to significantly increase the electrochemical performance of sensors based on $\mathrm{MoS}_{2}$-nanocomposites. The 2D $\mathrm{MoS}_{2} /$ metal-NS composites have the inherent properties of pure metal NSs and $\mathrm{MoS}_{2}$ nanosheets due to their synergistic effects, which makes the 2D $\mathrm{MoS}_{2} /$ metal-NS composites exhibit outstanding electrochemical properties and improved sensitivity [13]. Through studying Zhang's review paper ${ }^{[8]}$, one can find comprehensive insights into $\mathrm{MoS}_{2}$ nanosheets used as building blocks or supports for the preparation of $\mathrm{MoS}_{2}$ nanosheet-based composites with other materials.

## 5. Application of 2D $\mathrm{MoS}_{2}$-Based Nanocomposites

### 5.1. Electrochemical Biosensors

Recently, significant progress has been made in the development of electrochemical sensors and their application in point-of-care diagnostics, environmental studies, food safety, drug screening, and security. Electrochemical sensing has proven to be a simple analytical technique for detection of various chemicals and biological molecules due to intrinsic advantages, such as high sensitivity and selectivity, real-time measurements, low-cost instrumentation, and the potential for miniaturized and portable devices. A standard electrochemical sensor has three components, namely, a recognition component that specifically binds the target analyte, a transducer where specific reactions occur and a signal is produced, and an electronic component that converts the obtained signal to a response [113]. The operating mechanism of an electrochemical sensor involves the interaction of the target analyte with the electrode surface coated with a catalyst, and producing the desired change in the signal as a result of a redox reaction ${ }^{[114]}$. The reaction results in electrical, thermal, or optical output signals that can be used to investigate the nature of the analyte species [115]. The active sensing materials could be biological or chemical compounds that function as catalysts for sensing specific analytes [115]. Sensitivity and selectivity are essential variables in electrochemical sensors. Thus, the surface modifications of electrodes by immobilization of specific chemical or biological recognition elements is an efficient technique used for obtaining optimal binding of the target analyte. Commonly used biorecognition elements include enzymes [65], antibodies and antigens [66][92], aptamers [91], proteins [116] , etc., for high catalytic activity and excellent selectivity of the target analytes. Even so, the resulting signal from biorecognition molecules is not strong enough to achieve the ultra-sensitive detection of biomolecules required for early and rapid diagnosis of diseases [117]. Biorecognition molecules have therefore been integrated into nanomaterials to address this limitation, and to significantly improve the overall performance of the biosensor ${ }^{[118]}$. The 2D $\mathrm{MoS}_{2}$ nanosheets are among the nanomaterials that have become incredibly popular for sensing applications. They have been reported to be good electrode materials for electrochemical sensing; hence, there is an increasing number of publications addressing their integration into sensors [65][104]. Although 2D MoS 2 has low electrical conductivity compared to graphene due to its large band gap limiting its use as a pristine material, 2D MoS ${ }_{2}$ has tunable properties that depend on the crystal structure, nanosheet size, and surface defects [119][120][121], as already discussed in detail earlier. This gives researchers the opportunity to explore the electrocatalytic properties of non-functionalized 2D $\mathrm{MoS}_{2}$ for the detection of biological molecules. In a study presented by Wang et al. ${ }^{[122]}$, an electrochemical sensor based on non-functionalized $\mathrm{MoS}_{2}$ nanosheets was fabricated for detection of DNA. The reported results indicated that the bulk $\mathrm{MoS}_{2}$ had no electrocatalytic effect due to the low electronic conductivity that resulted from the poor interlayer electron transport. However, the exfoliated $\mathrm{MoS}_{2}$ nanosheets showed increased electrochemical activity with a decreased change in potential ( $\Delta \mathrm{Ep}$ ). The enhanced electrocatalytic activity of $\mathrm{MoS}_{2}$ nanosheets was discovered to be due to the anisotropic layered structure of $\mathrm{MoS}_{2}$ nanosheets, whose electronic structures and electrochemical activities are directly influenced by the layer thicknesses of the nanosheets ${ }^{[122]}$.

Moreover, Sha and co-workers explored the electrocatalytic properties of hydrothermally-grown non-functionalized $\mathrm{MoS}_{2}$ nanosheets on aluminum foil toward non-enzymatic detection of uric acid in human urine [123]. The group demonstrated a two-step successful growth of a few layered (< 4 layers) $\mathrm{MoS}_{2}$ nanosheets with a high ratio of 1 T phase MoS 2 than 2 H phase $\mathrm{MoS}_{2}$. This was not a typical sensor, in the sense that the aluminum foil was used as a sensor substrate, which contributed to the excellent selectivity and reproducibility, fast response time, and low limit of detection. The impressive sensing ability also resulted from a high proportion of the metallic 1 T phase $\mathrm{MoS}_{2}$, which provided excellent conductivity,
rapid electron transfer kinetics, and more exposed catalytic active sites arising from a significant number of surface defects ${ }^{[123]}$. The results of these two sensors are based on phenomena that describe the direct structure-dependent properties of $\mathrm{MoS}_{2}$. Changes in the interlayer coupling, the degree of quantum confinement, and symmetry elements result in major variations in the 2 D MoS 2 electronic structure compared to bulk [9][119][124][125]. The reduction in size of $\mathrm{MoS}_{2}$ offers abundant exposed edges, kinks, and corner atoms that induce additional chemical effects ${ }^{[126][127]}$. Moreover, the quantum size effects lead to changes in the valence band and the oxidation potential, thereby enabling catalytic activities not possible with the bulk band structure ${ }^{[126]}$ (Table 2).

Table 2. Electrochemical biosensors based on 2D-MoS ${ }_{2} /$ metal-NSs composites.

| Sensor | Analyte | Electrochemical Method | Linear Range | LOD | Ref. |
| :---: | :---: | :---: | :---: | :---: | :---: |
| Au-MoS ${ }_{2}$ /GCE |  | SWV | $1 \mathrm{nM}-10$ |  | [13] |
|  | ATP |  | mM | 0.32 nM |  |
|  |  |  |  |  |  |
|  | Thrombin |  | $0.01 \mathrm{nM}-10$ | 0.0014 nM |  |
|  |  |  | $\mu \mathrm{M}$ |  |  |
| GCE/Ni-MoS ${ }_{2} / \mathrm{Naf}$ | Glucose | Amperometry | 0-4 mM | 0.31 M | [35] |
| $\mathrm{MoS}_{2} / \mathrm{Au} / \mathrm{GOx}$ | Glucose | Amperometry | $\begin{gathered} 0.25-13.2 \\ \mathrm{mM} \end{gathered}$ | $0.042 \mu \mathrm{M}$ | [65] |
| $\mathrm{CuO} / \mathrm{MoS}_{2} / \mathrm{GCE}$ | Glucose | Amperometry | 35-800 $\mu \mathrm{M}$ | $0.017 \mu \mathrm{M}$ | [78] |
| $\mathrm{MoS}_{2}-\mathrm{Au} / \mathrm{Pt} @ \mathrm{GCE}$ | $\mathrm{H}_{2} \mathrm{O}_{2}$ | Amperometry | $\begin{gathered} 10 \mu \mathrm{M}- \\ 19.07 \mathrm{mM} \end{gathered}$ | $0.39 \mu \mathrm{M}$ | [80] |
| $\mathrm{Cu}-\mathrm{MoS}_{2} / \mathrm{GCE}$ |  | Amperometry | $\begin{gathered} 0.04-35.6 \\ \mu M \end{gathered}$ | $0.021 \mu \mathrm{M}$ | [81] |
|  | glucose |  | 1-70 $\mu \mathrm{M}$ | $0.32 \mu \mathrm{M}$ |  |
| ZnO/MoS $2 / \mathrm{GCE}$ | DNA | DPV | $\begin{gathered} 1.0 \mathrm{fM}-1.0 \\ \mu \mathrm{M} \end{gathered}$ | 0.66 fM | [82] |
|  |  | DPV | 20-300 |  | [83] |
|  | AA |  | $\mu \mathrm{mol} / \mathrm{L}$ | $3.0 \mu \mathrm{~mol} / \mathrm{L}$ |  |
| Au@MoS2/GCE | DA |  | 5-200 $\mu \mathrm{mol} / \mathrm{L}$ | $1.0 \mu \mathrm{~mol} / \mathrm{L}$ |  |
|  | UA |  | 20-400 | $5.0 \mu \mathrm{~mol} / \mathrm{L}$ |  |
|  |  |  | $\mu \mathrm{mol} / \mathrm{L}$ |  |  |
| $\mathrm{Au}-\mathrm{Pd} / \mathrm{MoS}_{2} / \mathrm{GCE}$ | $\mathrm{H}_{2} \mathrm{O}_{2}$ | DPV | $\begin{gathered} 0.8 \mu \mathrm{M}-10 \\ \mathrm{Mm} \end{gathered}$ | $0.16 \mu \mathrm{M}$ | [85] |
|  | Glucose | Amperometry | $0.5-20 \mathrm{mM}$ | 0.40 mM |  |
| Pt-MoS ${ }_{2} / \mathrm{GCE}$ | $\mathrm{H}_{2} \mathrm{O}_{2}$ | Amperometry | $\begin{gathered} 0.004-48.5 \\ \mathrm{mM} \end{gathered}$ | 0.001 mM | [87] |


| Sensor | Analyte | Electrochemical <br> Method | Linear <br> Range | LOD | Ref. |
| :---: | :---: | :---: | :---: | :---: | :---: |
|  | DA |  | 0.5-250 $\mu \mathrm{M}$ | $0.1 \mu \mathrm{M}$ |  |
| PtNi@MoS ${ }_{2}$ /GCE | UA | DPV | $\begin{gathered} 0.5-1800 \\ \mu \mathrm{M} \end{gathered}$ | $0.1 \mu \mathrm{M}$ | [88] |
| $\mathrm{Cu}_{2} \mathrm{O} / \mathrm{MoS}_{2} / \mathrm{GCE}$ | Glucose | Amperometry | $\begin{gathered} 0.01-4.0 \\ \mathrm{mM} \end{gathered}$ | $1.0 \mu \mathrm{M}$ | [89] |
| PtW/Mos $/$ /GCE | $\mathrm{H}_{2} \mathrm{O}_{2}$ | Chronoamperometry | $\begin{gathered} 1 \mu \mathrm{M}-0.2 \\ \mathrm{mM} \end{gathered}$ | 5 nM | [90] |
| Pd/PDDA-G-MoS ${ }_{2} / \mathrm{GCE}$ | TB | DPV | $\begin{gathered} 0.0001-40 \\ \mathrm{nM} \end{gathered}$ | 0.062 pM | [91] |
| PtNPs@MoS ${ }_{2}$ GCE | DA <br> UA | DPV | 0.5-150 <br> $\mu \mathrm{mol} / \mathrm{L}$ <br> 5-1000 $\mu \mathrm{mol} / \mathrm{L}$ | 0.12 <br> $\mu \mathrm{mol} / \mathrm{L}$ <br> $0.8 \mu \mathrm{~mol} / \mathrm{L}$ | [93] |
| Ag@ $\mathrm{MoS}_{2} / \mathrm{GCE}$ | DA | DPV | 1-500 $\mu \mathrm{M}$ | $0.2 \mu \mathrm{M}$ | [94] |
| $\mathrm{MoS}_{2}$-CPtNPs/GCe |  | DPV | 5-200 $\mu \mathrm{M}$ | $0.39 \mu \mathrm{M}$ | [95] |
|  | UA |  | $20-500 \mu \mathrm{M}$ | $1.8 \mu \mathrm{M}$ |  |
| Laminin/Au-Pd-Pt/MoS2/SPCE | $\mathrm{H}_{2} \mathrm{O}_{2}$ | Amperometry | 1-100 nM | 0.3 nM | [96] |
| $\mathrm{NiO} / \mathrm{MoS}_{2} / \mathrm{GCE}$ | Glucose | Amperometry | 0.01-10 mM | $1.62 \mu \mathrm{M}$ | [99] |
| GCE/Cu-MoS2/Nafion | Glucose | Amperometry | 0-4 mM | - | [104] |
| $\mathrm{NiCo}_{2} \mathrm{O}_{4}-\mathrm{MoS}_{2} /$ chitosan/GCE | Glucose | Amperometry | $\begin{gathered} 0.0007- \\ 13.78 \mathrm{mM} \end{gathered}$ | $0.23 \mu \mathrm{M}$ | [105] |
| $\mathrm{MoS}_{2}$-PPY-AuNPs/GCE | Glucose | DPV | $0.1-80 \mathrm{nM}$ | 0.08 nM | [128] |
| AuNPs@Mos ${ }_{2} / \mathrm{GCE}$ | miRNA-21 | DPV | $10 \mathrm{fM}-1 \mathrm{nM}$ | 0.78 fM | [129] |
| Chox/MoS ${ }_{2}$-AuNPs/GCE | Cholesterol | Amperometry | $0.5-48 \mu \mathrm{M}$ | $\begin{gathered} 0.26 \pm \\ 0.015 \mu \mathrm{M} \end{gathered}$ | [130] |
| $\mathrm{MoS}_{2}$-Au-PEI-hemin | Clenbuterol (CLB) | DPV | $10 \mathrm{ng} / \mathrm{mL}-2$ $\mu \mathrm{g} / \mathrm{mL}$ | $\begin{gathered} 1.92 \\ \mathrm{ng} / \mathrm{mL} \end{gathered}$ | [131] |
| NF/AuNPs/CuO-MoS ${ }_{2}$ | Glucose | Chronoamperometry | $\begin{gathered} 0.5 \mu \mathrm{M}-5.67 \\ \mathrm{mM} \end{gathered}$ | $0.5 \mu \mathrm{M}$ | [132] |


| Sensor | Analyte | Electrochemical Method | Linear Range | LOD | Ref. |
| :---: | :---: | :---: | :---: | :---: | :---: |
| MCH/dsDNA/MoS ${ }_{2}$-AuNPs/GCE | T4 polynucleotide kinase <br> (PNK) | SWV | $\begin{gathered} 0.001-10 \\ \mathrm{U} / \mathrm{mL} \end{gathered}$ | $\begin{gathered} 2.18 \times \\ 10^{-4} \mathrm{U} / \mathrm{mL} \end{gathered}$ | [133] |
| miRNA/MCH/SH-RNA/AuNPs$\mathrm{MoS}_{2} / \mathrm{FTO}$ | MicroRNA-155 | DPV | $1 \mathrm{fM}-10 \mathrm{nM}$ | 0.32 fM | [134] |

ATP; triphosphate, DA; dopamine, DNA; deoxyribonucleic acid, DPV; differential pulse voltammetry, SWV; square Wave Voltammetry, PDDA-G; poly(diallyldimethylammonium chloride)-graphene, TB; thrombin, CPtNPs; Clover-like platinum nanoparticle, PPY; polypyrrole, miRNA-21; microribonucleic acid-21, Chox; cholesterol oxidase, NF; Nafion, MCH; 6mercaptohexanol, FTO; fluorine doped tin oxide.

### 5.2. Electrochemical Immunosensors

Electrochemical immunosensors (EIs) have received considerable attention due to their high sensitivity, low cost, simple instrumentation and operation, and inherent miniaturization [135][136][137]. They show great potential in the next-generation of point-of-care (POC) diagnostics for early detection and monitoring of diseases. An electrochemical immunosensor is a type of biosensor used to detect the specific antigen-antibody recognition; it quantitatively measures the resulting electrochemical signal [66][103]. The basic immunosensor concept is to load the antibody onto the electrode surface. Following the specific antigen-antibody binding, the electron transfer rate between the electrode surface and the solution interface changes, resulting in the production of membrane potential and change of the current response, which directly reflects the concentration of the targeted antigen ${ }^{[103]}$. This type of immunoassay sandwich is a common format of an immunosensor. Countless efforts have been made into the advancements of Els to improve performance by achieving high sensitivity. This includes the use of different nanomaterials, such as layered nanomaterials $\frac{[138]}{}$, metal and metal oxides NSs ${ }^{[139][140]}$, and carbon-based nanomaterials ${ }^{[141]}$ to amplify the El signals. Due to excellent catalytic activity and biocompatibility, $\mathrm{MoS}_{2}$ functionalized with metal or metal oxide NSs has recently been introduced among nanocomposites used in the construction of immunosensors (Table 3).

Table 3. Electrochemical immunosensors based on 2D-MoS $2 /$ metal-NSs composites.

| Sensor | Analyte | Electrochemical <br> Method | Linear Range | LOD | Ref. |
| :---: | :---: | :---: | :---: | :---: | :---: |
| $\mathrm{MoS}_{2} @ \mathrm{Cu}_{2} \mathrm{O}-\mathrm{Pt} / \mathrm{Ab}_{2}$ | hepatitis $B$ antigen | Amperometry | $0.5 \mathrm{pg} / \mathrm{mL}-$ $200 \mathrm{ng} / \mathrm{mL}$ | $\begin{gathered} 0.15 \\ \mathrm{pg} / \mathrm{mL} \end{gathered}$ | [66] |
| BSA/anti-HBs/PtPd NCs@MoS 2 GCE | Hepatitis B antigen | DPV | $32 \mathrm{fg} / \mathrm{mL}-$ <br> $100 \mathrm{ng} / \mathrm{mL}$ | $\begin{array}{r} 10.2 \\ \mathrm{fg} / \mathrm{mL} \end{array}$ | [92] |
| $\mathrm{MoS}_{2} @ \mathrm{Cu}_{2} \mathrm{O}-\mathrm{Au}-\mathrm{Ab}_{2}$ | Alpha fetoprotein (AFP) | Amperometry | $0.1 \mathrm{pg} / \mathrm{mL}-$ $50 \mathrm{ng} / \mathrm{mL}$ | 0.037 <br> pg/mL | [98] |
| $\mathrm{Ab}_{2}-\mathrm{Ag} / \mathrm{MoS}_{2} @ \mathrm{Fe}_{3} \mathrm{O}_{4} / \mathrm{MGCE}$ | carcinoembryonic antigen (CEA) | DPV | $0.0001-20$ <br> ng/mL | $\begin{gathered} 0.03 \\ \mathrm{pg} / \mathrm{mL} \end{gathered}$ | [103] |
| HRP/HRP-anti-CEA/MoS $2^{-}$ AuNPs | carcinoembryonic antigen (CEA) | DPV | $10 \mathrm{fg} / \mathrm{mL}-1$ $\mathrm{ng} / \mathrm{mL}$ | $\begin{gathered} 1.2 \\ \mathrm{fg} / \mathrm{mL} \end{gathered}$ | [142] |


| Sensor | Analyte | Electrochemical <br> Method | Linear Range | LOD | Ref. |
| :---: | :---: | :---: | :---: | :---: | :---: |
| $\mathrm{GCE} / \mathrm{MoS}_{2}-\mathrm{Au}-\mathrm{Ab}_{1}$ | CEA | DPV | $1 \mathrm{pg} / \mathrm{mL}-50$ $\mathrm{ng} / \mathrm{mL}$ | $0.27$ <br> pg/mL | [143] |
| Pd NPs@Mos ${ }_{2} /$ NiCo | Procalcitonin | Chronoamperometry | $0.001-50$ <br> ng/mL | $\begin{gathered} 0.36 \\ \mathrm{pg} / \mathrm{mL} \end{gathered}$ | [144] |
| $\mathrm{Au}-\mathrm{MoS}_{2} / \mathrm{ITO}$ | Triiodothyronine ( $\mathrm{T}_{3}$ ) | EIS | $\begin{gathered} 0.01-100 \\ \mathrm{ng} / \mathrm{mL} \end{gathered}$ | $\begin{gathered} 2.5 \\ \mathrm{pg} / \mathrm{mL} \end{gathered}$ | [145] |
| $\mathrm{Cu}-\mathrm{MoS}_{2} / \mathrm{GCE}$ | 3-phenoxybenzoic acid (3-PBA), | EIS | 0-6 $\mu \mathrm{g} / \mathrm{mL}$ | $3.8 \mu \mathrm{M}$ | [146] |
| Tac/BSA/Ab/PS-AuNRs@L-Cys-MoS2/GCE | Tacrolimus (Tac) | DPV | $1.0-30$ <br> $\mathrm{ng} / \mathrm{mL}$ | $\begin{gathered} 0.17 \\ \mathrm{ng} / \mathrm{mL} \end{gathered}$ | [147] |

Ab $_{1}$; primary antibody, $\mathbf{A b}_{2}$; secondary antibody, $\mathbf{H B s} \mathbf{A g}$; Hepatitis B surface antigen, BSA; bovine serum albumin, ITO; indium tin oxide, L-Cys; L-cysteine, PS-AuNRs; polystyrene-gold nanorods.

### 5.3. Photoelectrochemical Sensors

PEC sensing has experienced rapid growth as an important branch of electrochemical detection. PEC analysis, due to high sensitivity, fast detection, and low background current, has been wildly adopted in bioanalytical chemistry as a newly emerging and continuously developing analytical method [148]. The standard PEC sensor uses light as an excitation source and photocurrent as a signal identifier. Photoactive materials are immobilized on the electrode surface as a photo-to-current converter to generate photocurrent signals under light irradiation. The photo-to-current conversion results from the electron excitation and subsequent charge transfer of a material after absorbing photons [149][150]. The total separation of the source of excitation (light) and detection signal (photocurrent) enables this method to have the advantages of a low background signal and high sensitivity to recognize target analytes in complex samples as opposed to traditional optical methods [151][152]. Photoactive materials play critical roles in PEC systems; The PEC sensor output depends on the properties of these materials since they can influence the sensitivity and selectivity of the PEC sensor. Different photoactive materials, such as metal-based NPs ${ }^{[152][153]}$, quantum dots (QDs) ${ }^{[154]}$, carbon-based nanomaterials ${ }^{[155]}$, layered nanomaterials ${ }^{[148]}$, etc., have been studied in PEC application to fulfil specific demands. Furthermore, biological molecules, such as enzymes, antibodies, nucleic acids, etc., are also important for specific recognition in PEC biosensors and immunosensors. Amongst the photoactive materials, 2D $\mathrm{MoS}_{2}$ is the latest emerging material for use in PEC biosensors and immunosensors because of its special optical properties. Hun et al. ${ }^{[148]}$ developed a PEC sensor based on a single-layer $\mathrm{MoS}_{2}$ nanosheet for detection of dopamine (DA). The fabricated sensor showed an incredible photocurrent response, which demonstrated that PEC activity was enhanced by the single-layer $\mathrm{MoS}_{2}$. That is because single-layer $\mathrm{MoS}_{2}$ exhibits exceptional semiconducting electronic properties as well as a good photocurrent with a sensitive photoresponse compared to multilayer $\mathrm{MoS}_{2}$. The photocurrent proportionally increased with the concentration of DA in the range of 0.01 nM to $10 \mu \mathrm{M}$ with a detection limit of 0.0023 nM . Even though different photoactive materials are available for the fabrication of PEC sensors, due to the limitations of a single material, it is difficult to achieve the everincreasing demands for detection. However, any drawbacks of the pristine photoactive material, such as $\mathrm{MoS}_{2}$, can be resolved by flexible nanocomposites with the tailor-made structure and composition ${ }^{[156]}$. Additionally, certain biological molecules, such as cells and DNA, can produce photocurrents by themselves. Thus, the interactions between these biomolecules and other photoactive materials can be examined through the change of photocurrents ${ }^{[149]}$. In general, PEC biosensing or immunosensing refers to the effect on the photocurrent signal of the interaction between the biorecognition molecule and the analyte, which involves the charge and energy transfer of the PEC reaction between the electron donor/acceptor and the photoactive material during light irradiation ${ }^{[151]}$ (Table 4).

Table 4. Photoelectrochemical sensors based on 2D-MoS ${ }_{2} /$ metal-NSs composites.

| Sensor | Analyte | Linear Range | LOD | Ref. |
| :---: | :---: | :---: | :---: | :---: |
| TTR/AuCNs/Mos ${ }_{2} / \mathrm{GCE}$ | Tetrabromobisphenol A | $0.1 \mathrm{nM}-1.0 \mu \mathrm{M}$. | 0.045 nM | [76] |
| $\mathrm{BSA}\|a p t a m e r\| \mathrm{TiO}_{2}-\mathrm{MoS}_{2}{ }^{-}$ AuNP\|ITO | kanamycin | $0.2 \mathrm{nM}-450 \mathrm{nM}$ | 0.05 nM | [86] |
| GOx\| $\mathrm{MoS}_{2}-\mathrm{TiO}_{2}$ \|ITO | Glucose | $0.1-10.5 \mathrm{mM}$ | 0.015 mM | [102] |
| $\mathrm{MoS}_{2}$-ZnO\|ITO | Propyl gallate | $\begin{gathered} 0.1249-1643 \\ \mu \mathrm{~mol} / \mathrm{L} \end{gathered}$ | $\begin{gathered} 1.2 \times 10^{-8} \\ \mathrm{~mol} / \mathrm{L} \end{gathered}$ | [157] |
| $\mathrm{Au} / \mathrm{MoS}_{2} / \mathrm{TiO}_{2}$ | Glucose | 5-1000 $\mu \mathrm{M}$ | 1.3 nM | [158] |
| Pro-GRP-MIP/AuNPs/2D$\mathrm{MoS}_{2} / \mathrm{GCE}$ | Pro-gastrin-releasing peptide (ProGRP) | 0.02-5 ng/mL | $0.0032 \mathrm{ng} / \mathrm{mL}$ | [159] |
| $\mathrm{Au}-\mathrm{MoS}_{2} / \mathrm{FTO}$ | anti-human IgG | $41.7 \mathrm{nM}-4.17 \mu \mathrm{M}$ | 4.17 nM | [160] |
| ITO/MTiO ${ }_{2}$-AuNPs-MoS ${ }_{2}$-GOx | Glucose | $0.004-1.75 \mathrm{mM}$ | $1.2 \mu \mathrm{M}$ | [161] |
| biotin DNA/MoS ${ }_{2}$-AuNPs/ITO | miRNA | $10 \mathrm{fM}-1 \mathrm{nM}$ | 4.21 fM | [162] |

MIP; molecularly imprinted polymer.

### 5.4. Electrochemiluminescence Sensors

ECL detection is commonly used in the various applications for biosensors. This technique combines the advantages of electrochemistry and chemiluminescence in resolving single sensing limitations, making it more desirable for precise detection and enhancement of a sensor's sensitivity and specificity. ECL refers to the light emission from an excited state produced by the electron transfer reaction between species at electrode surfaces following electrochemical reactions [163]. ECL detection consists of measuring photon output and, thus, the light intensity emitted in the solution during an electrochemical reaction. The light intensity is therefore directly proportional to the concentration of one or more of the reactants involved in the electrochemical reaction ${ }^{[164]}$. ECL has many advantages over photoluminescence, such as simplicity, high sensitivity, and rapidity response, and does not require the use of an external light source, leading to a high luminescent purity and a low optical background noise ${ }^{[163][165]}$. Furthermore, the ECL emission light can be initiated and controlled with high reproducibility and accuracy by alternating the applied potential [163] 166 . Unfortunately, one of the drawbacks of the ECL method is the possibility of electrode fouling, which can result in poor reproducibility. While this could sometimes be a concern in the presence of complex samples, this effect can be avoided by frequent electrochemical cleaning of the electrodes. Moreover, new electrode materials that are less prone to fouling, such as boron-doped diamond and disposable screen-printed electrodes, have been introduced and have become more popular [167]. The continuous development of nanomaterials offers new opportunities for conventional ECL research, mainly owing to their large surface area, abundant active sites, and distinctive optical properties. Nanomaterials with good electron transfer capabilities, such as 2 D nanomaterials ${ }^{[168]}$, quantum dots ${ }^{[169]}$, and metal nanoparticles ${ }^{[170]}$, have also been implemented to improve the efficiency of ECL sensors. Among these nanomaterials, the use of 2 D MoS 2 in ECL sensors is not so prominent. This is attributable to the fact that 2 D MoS 2 is not capable of generating electrochemiluminescence; however, it serves other functions, such as acting as a robust substrate or amplifying the signal ${ }^{[171]}$. $\mathrm{MoS}_{2}$ can also be used as a carrier for other nanomaterials with structural and compositional benefits to improve the ECL performance.

Table 5 summarizes the performances of a few ECL sensors that have been reported based on 2D-MoS $2 /$ metal-NSs composites.

Table 5. Electrochemiluminescence sensors based on 2D-MoS $2 /$ metal-NSs composites.

| Sensor | Analyte | Linear Range | LOD | Ref. |
| :---: | :---: | :---: | :---: | :---: |
| luminol-Au@BSA-Ab $2 / \mathrm{AFP} / \mathrm{BSAT} / \mathrm{Ab}_{1} / \mathrm{Chi}^{-} / \mathrm{MoS}_{2^{-}}$ PEI-Au/GCE. | Alpha fetal protein (AFP), | 0.0001-200.0 $\mathrm{ng} / \mathrm{mL}$ | $\begin{gathered} 1.0 \times 10^{-5} \\ \mathrm{ng} / \mathrm{mL} \end{gathered}$ | [77] |
| BSA/Ab2/ABEI-Cys/Au-Pd-Pt/MoS ${ }_{2}$ | cystatin C (CYSC) | $\begin{gathered} 1.0 \mathrm{fg} / \mathrm{mL}-5.0 \\ \mathrm{ng} / \mathrm{mL} \end{gathered}$ | $0.35 \mathrm{fg} / \mathrm{mL}$ | [172] |
| QDs-Apt2/PDGF-BB/Apt1/MoS ${ }_{2}$-AuNPs/GCE | platelet-derived growth factor-BB | 0.01-100 pmol/L | 1.1 fmol/L | [173] |

ABEI; N-(aminobutyl)-N-(ethylisoluminol), PDGF-BB; platelet-derived growth factor BB, QDs; quantum dots.
Focusing on the application of 2D $\mathrm{MoS}_{2}$-metal NS composites on various electrochemical sensors, it can be inferred that $2 \mathrm{D} \mathrm{MoS}_{2}$ represents a new class of highly electroactive materials. The large surface areas of 2 D MoS 2 nanosheets enable surface functionalization with a variety of nanomaterials, providing new opportunities for 2D material-based functional devices. The effective support structure of $\mathrm{MoS}_{2}$ nanosheets allows for a good distribution of metal-based NSs, resulting in a synergistic effect that enhances catalytic efficiency and conductivity over a pristine $\mathrm{MoS}_{2}$. Furthermore, 2D$\mathrm{MoS}_{2}$ /metal-NS composites provide a convenient platform for the immobilization of biorecognition molecules and enhance electron transfer, resulting in faster reaction times as well as increased sensitivity and selectivity. Therefore, 2D MoS ${ }_{2}-\mathrm{NSs}^{2}$ composites are thought to have considerable potential as effective electrocatalysts for the production of highly responsive electrochemical sensors with low limits of detection.

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