# **N-Type Metal Oxide Semiconductor: Materials and Their Environmental Applications**

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#### Scopus Author ID 41762253400 Received: 30.09.2022; Accepted: 4.11.2022; Published: 4.02.2023

**Abstract:** Semiconductors are the basic building block and basic elements of modern electrical devices and machines. N-type metal oxide semiconductor (MOS) is particularly interesting due to their unique properties and wide applications. Because of their enormous applications and importance, semiconductors are believed to play a major role in facilitating modern life. Medicine, agriculture, mechanics, nuclear energy, biotechnology, communications, and data operations are among the areas that benefit most from the application of semiconductors. Hence, this review attempts to summarize the important features of semiconductors in general and MOS nanoparticles in particular, their structure and properties. The applications of MOS and the thin-film transistor are summarized with emphasis on their applications as photocatalysts for bioremediation, solar and hydrogen cells, and sensor devices.

# **Keywords:** semiconductor; N-type; metal oxide nanoparticles; sensors; biosensors; solar cells; hydrogen production.

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# 1. Introduction

According to their electrical properties, materials can be classified into three groups: conductors, semiconductors, and insulators. Semiconductors are a large class of crystalline solids with electrical conductivity and resistivity behavior between conductors and nonconductors (insulators). As its structure contains very few free electrons (from added impurities), it acts at room temperature as an insulator. An important property of semiconductors is the conductivity that increases in response to an increase in temperature, unlike metals, whose conductivity decreases with increasing temperature. Semiconductors are the basic "building block" and "key elements" for modern electrical devices and machines. They are employed in almost all electronic industries, including transistors, integrated circuits, computers, solar cells, and light-emitting diodes (LEDs). The applications and importance of semiconductors are clear in cable networking, wireless networking, satellites, optical sensors, and gas sensors. Due to their vast applications and importance, semiconductors are thought to play a main role in making modern life easier. Medicine, agriculture, mechanics, nuclear energy, biotechnology, communications, and data operations are among the areas that benefit most from the application of semiconductors [1-3]. Semiconductors are generally applied for manufacturing many devices like diodes, transistors, "metal oxide silicon field-effect transistors" (MOSFET), "junction-gate field-effect transistors" (JFET), unijunction transistors

(UJT), sensors, etc. Sensor-based smart systems are among the most important applications used daily in almost all human activities [4]. Sensors represent a "fast, sensitive, selective, reliable, and low-cost" analytical tool that benefits all fields of life. Therefore, the role of semiconductors, particularly N-type metal oxide semiconductors in sensors, is very important.

Hence, in this review, semiconductors are addressed in terms of concept, types, and some of their applications. The synthesis and applications of N-type metal oxide semiconductors, particularly gas sensors, are also discussed. In addition, flexible oxide semiconductor TFTs, the dielectric fabrication process, and the evaluation of sensing (gas sensing) were also included during this review.

#### 2. History of Semiconductors

Semiconductors recognition and understanding were started throughout the mid-19th decade during the experiments on the "electrical properties" of materials. Michael Faraday discovered that, unlike metals, the resistance of silver sulfide increases with increasing temperature. Also, Willoughby Smith reported a resistance decrease in selenium resistors when exhibited decreasing when exposed to light. Similar "time-temperature coefficient of resistance", rectification, and "light-sensitivity" observations for different materials lead to understanding the semiconductor's nature and importance. Arthur Schuster found that the copper oxide layer on wires has corrective properties that disappear when the wires are cleaned. The theories regarding electron-based conduction in solids and electron discovery by J.J. Thomson were developed in the late nineteenth century. The classification of conductors (metals) and "variable conductors" was specified by Johan Koenigsberger in 1914, while the term semiconductor or "Halbleiter" was mentioned earlier in 1910 in a doctoral dissertation by Koenigsberger's student Josef Weiss [1,5,6]. However, according to Łukasiak and Jakubowski [7], the term "semiconductor" "was used for the first time by Alessandro Volta in 1782".

Moreover, they noted that the "conduction and rectification in metal sulfides probed with a metal point (whisker)" was observed in 1874 by Karl Ferdinand Braun. Such studies and funding were considered later as a starting point for developing and detecting radio and microwave radiation in WWII radar systems. The point-contact and junction transistor were invented by "Bardeen and Brattain" and "Shockley" in 1947 and 1948, respectively, ushering in the era of the transistor. Later, in 1956, Shockley, Bardeen, and Brattain were awarded the Nobel Prize (jointly) in Physics due to their efforts, development, and achievements in semiconductor and transistor research. A major remark in semiconductor history occurred with the invention of "integrated circuits" (ICs) in the US in 1957, which became smaller in size and wider in different electric applications with time [5,8]. The metal–oxide–semiconductor (MOS) was utilized in the late 1950s in the so-called "MOS field-effect transistor". MOS field-effect transistors are mass-produced for various uses, including computers, communications technology (such as smartphones), and electronics such as gas and light sensors [6,9]. Since this time, the development in the field of semiconductors has increased and covered every corner of our life.

## 3. Insulators, Conductors, and Semiconductors

Metals such as copper, aluminum, silver, and gold are good conductors because their atoms have "loosely" valence electron(s) bound to the atom therefore they can be "available to carry current". At the same time, materials such as glass, rubber, and ceramics are considered

insulators because they prevent electrical conduction due to their high resistance. On the other hand, materials such as silicon, germanium, or gallium arsenide have low free electrons that allow electrical conductivity under certain conditions "but not others" [2]. This property of semiconductors makes them neither a good conductor nor a good insulator". This unique "inbetween" electrical conductivity of semiconductors enables "passing current more easily in one direction than the other, showing variable resistance, and having sensitivity to light or heat". The conductivity of semiconductors could be increased in controlled degree by adding some impurities of metals ("doping") into its crystal structure [10,11]. However, there is another important feature of semiconductors, the conductivity of which increases with increasing temperature (unlike metals whose conductivity decreases with increasing temperature) [12]. Silicon is one of the most important, abundant, and applied semiconductor materials. It is mainly used in the manufacture of integrated electronic circuits as well as in solar cells and laser diodes. Gallium arsenide, germanium, and indium antimonite are common semiconductors used in "low-noise, high-gain, low signal amplification". In addition, metal oxides (such as SnO<sub>2</sub>, ZnO, WO<sub>3</sub>, and TiO<sub>2</sub>) as ionic solids and conventional nonstoichiometric oxides are promising semiconductors in a wide range of applications [2,13].

## 4. Classification of Semiconductors

Semiconductor materials can be broadly classified based on the energy gap into intrinsic and extrinsic semiconductors, as illustrated in the following flow chart (Figure 1).



Figure 1. Flowchart of different semiconductors types.

## 4.1. Intrinsic semiconductors.

This type of semiconductor is made from a pure material without any impurities. In intrinsic semiconductors, the number of holes is equal to the number of electrons in the conduction band. Pure germanium and silicon are examples of such semiconductors where that have small "forbidden energy gaps" (0.72 and 1.1 eV, respectively). In an intrinsic semiconductor, the available energy is sufficient even at room temperature to enable the valence electrons to "jump through the conduction band". This type of semiconductor uses is very limited due to electrical conductivity at room temperature and above [1].

#### 4.2. Extrinsic semiconductors.

In an extrinsic semiconductor, the pure state of the semiconductor material changes by adding impurities in very minute quantities (the ratio does not exceed 0.01 ppm). These added impurities are known as "dopant(s) or doping agent(s)". The size of the semiconductor and dopant atoms should be the same, and the amount of dopant should not change the " lattice structure of the semiconductor". Doping materials are chosen to be either pentavalent or trivalent (having five or three electrons, respectively, in the valence band). According to the valent of doping material, extrinsic semiconductors are classified into two types, i.e., n-type and p-type [1,3].

#### 4.3. N-type semiconductor.

The so-called n-type semiconductor occurs when the semiconductor materials such as Si and Ge are doped with pentavalent elements. In this case, the semiconductor atoms (Si and Ge) belong to the fourth group in the periodic table, and the doping atoms belong to the fifth, ensuring a close size between the atoms. Moreover, when Si atoms are doped with a pentavalent atom, so four of its electrons will bond with four Si atoms while the fifth electron will remain loosely (to its doping atom). This loose electron requires very small ionization energy to be free and move at room temperature. It is important to mention that the 'n' syllable in the n-type name denotes the negative charge on the loose electron due to doping. These electrons in n-type are considered the "majority current carriers"; however, a few "holes" (or the so-called minority carriers) are also created thermally due to electron-hole pairs generation (Figure 2). The pentavalent atoms used for doping semiconductors include arsenic, phosphorous, antimony, and bismuth [1,3,11,14].



#### 4.4. P-type semiconductor.

P-type refers to the "positive charge" that occurred due to the addition of impurity atoms with trivalent to silicon. In this case, the trivalent atom bonds with four neighbor silicon atoms, so a hole (with a positive charge) results for each trivalent doping atom. The trivalent atom is referred to as an acceptor atom because it can take an electron. The number of "holes" formed equals the number of trivalent atoms added to silicon, and these holes are considered the majority carriers. However, a few conduction-band electrons are created in addition to holes



due to the thermal generation of electron-hole pairs (minority carriers) (Figure 3). The most applied trivalent dopants in p-type semiconductors are boron, indium, and gallium [3,15].

Figure 3. P-type semiconductors.

#### 4.5. The p-n junction.

When p- and n-type junctions occur within a single semiconductor crystal, a p-n junction is formed where "majority carriers" are located in the "holes" on the p-side and on the "electrons" of the n-side. There is a region near the junction without free charge carriers called "the depletion layer" which behaves like an insulator. The current will flow forward only in one direction (biased) (basic diode). The "non-reversing behavior" occurs due to the "nature of the charge transport process in the two types of materials". The p-n junctions are used as a rectifier (in electric circuits) and a voltage-controlled oscillator (in varactors) [16,17].

## 5. Semiconductor Materials

The semiconductors could be formed from a single element such as Si or Ge, or it could be a mixture of two or more semiconductors elements. Silicon is the most used material in semiconductor manufacturing, followed by gallium arsenide and germanium. In the periodic table, those common semiconductor elements are located near the so-called "metalloid staircase". As mentioned earlier, semiconductor materials are doped with impurities under "precise conditions" in order to convert the "Intrinsic" into "Extrinsic semiconductor". The nature of the impurities (dopants) will determine the type of produced extrinsic semiconductor type, i.e., p- or n-type.

The conservative buffer layer construction procedure on a glass substrate is losing consideration due to its high-water vapor transmission rate (WVTR) of transporter glass. Moreover, the organic-based flexible substrates have a poor WVTR [18,19], and the layer of buffer displays a very important role in a stable thin-film transistor (TFT). Among the used materials, silicon nitride (SiN<sub>x</sub>), silicon oxide (SiO<sub>2</sub>), and Al<sub>2</sub>O<sub>3</sub> are generally used for the flexible substrate buffer layer [20,21]. SiNx and SiO<sub>2</sub> are made-up through the plasma-enhanced chemical vapor deposition (PECVD) course and are already being used for the Si and oxide TFT passivation layers, and SiNx is better in the water encapsulation goods because it has a higher WVTR compared with SiO<sub>2</sub>. However, the PECVD development for SiNx has an extremely high hydrogen (H<sub>2</sub>) concentration (~25%) and hydrogen diffusion, which may degrade the TFT performance. In the case of Al<sub>2</sub>O<sub>3</sub>, it is usually made up through ALD and

has a low concentration of  $H_2$  with a high WVTR compared to silicon-based thin films [22-23].

# 6. The Technology of Metal-Oxide Semiconductors

Usually, Oxides are thought to be insulators. However, some metal oxides have the properties of semiconductors [24]. Metal oxide semiconductors (MOS) have electronic chargetransfer properties that make them a "unique class of semiconductors" compared to conventional covalent materials, such as Si [25]. MOS are conventional non-stoichiometric oxides with a cation metallic moiety (i.e., Zn, Cu, and Ni) strongly bound by an ionic bond with negative oxygen ions. This class of materials shows unique sensitivity toward electrical conductivity and "superior thermal/chemical stability" compared with free-metal oxides [26]. Because of their characteristics, MOSs have a wide range of applications that require films with large surface area, low processing temperature, flexibility, and particularly low cost [13]. Conventional silicon-based transistors have some limitations and difficulties, i.e., "limited form-factor, the difficulty of the large-area process, and high-cost complex-to-fabricate process". Hence, the advantages of metal-oxide-based transistors make them a suitable candidate for the next generation of thin-film transistors (TFTs) for foldable, flexible, and stretchable displays and other electronic products [27]. The MOS could be either p-type or ntype polycrystalline silicon metals. Tin monoxide (SnO) and CuxO are examples of common p-type oxide semiconductors. At the same time, zinc oxide (ZnO), indium gallium zinc oxide (IGZO), and tin dioxide (SnO<sub>2</sub>) are of most popular n-type oxide semiconductors [13]. Figure (4) represents a simple composition of metal oxide semiconductors.



Figure 4. A simple composition of metal oxide semiconductors.

# 7. Preparation and Synthesis of Metal Oxide Nanoparticles

MOS can be synthesized by various methods, including hydrothermal synthesis, coprecipitation, spray pyrolysis, and sequential ionic layer adsorption and reactivity (SILAR). The use of high vapor pressures and temperatures to crystallize compounds from their aqueous solutions is known as hydrothermal synthesis or "hydrothermal techniques". Metal precipitation from its salt in a solvent is the basis of the co-precipitation process. It's a quick and easy way to make mono-dispersed nanoparticles. The metal precursor solution (in the form of aerosols) is sprayed on a hot substrate via a nozzle in the spray pyrolysis technique. Flow rate, temperature, additives, and the concentration of the metal's precursor solution might be used to control synthesized metal oxides' particle size and shape. The SILAR approach, on the other hand, "relies on substrate immersion to establish cations and anions". It's used to make various thin-film materials and deposit them uniformly onto the substrate surface(s) at room temperature [28]. Metal oxides (including rare-earth-based nanomaterials) are often synthesized using hydrothermal, solvothermal/hydrothermal, sol-gel, and co-precipitation techniques [29,30]. Many ways may be used to produce MOS (Figure 5 & Table 1).



Figure 5. Chart of Synthetic routes for the preparation of metal oxide nanomaterials.

Table 1. Comparison of physical, chemical, and biological methods for nanomaterials synthesis.									
Synthesis method	Description	Advantages	Disadvantages	Notes	Ref.				
Physical	<ul> <li>Through mechanical pressure, high-energy radiations, and thermal or electrical energy.</li> <li>Common methods: laser pyrolysis, spray pyrolysis, high-energy ball milling, physical vapor deposition, melt mixing, laser ablation, sputter deposition, electric arc deposition, ion implantation, etc.</li> </ul>	<ul> <li>Quick process.</li> <li>Devoid of solvents (free from contamination through solvent)</li> <li>Uniform distribution of nanoparticles.</li> </ul>	<ul> <li>Less feasible: during the synthesis processes, a profuse amount of waste is produced.</li> <li>The final particles of the product do not have an orderly shape.</li> <li>Energy-intensive</li> <li>Need special devices</li> </ul>	- Generally based on the top-down approach.					
Chemical	<ul> <li>Mainly based on the chemical reduction of ions in their aqueous solutions by organic and inorganic reducing agents.</li> <li>For the synthesis of nanomaterials, several chemical methods are also used, such as hydrothermal synthesis, chemical vapor synthesis, sol-gel method, chemical vapor deposition, microemulsion technique, and polyol synthesis.</li> </ul>	<ul> <li>Quick process.</li> <li>Allow synthesis of nanoparticles in large quantities.</li> <li>Possibility of controlling particle size.</li> <li>These methods are relatively simple, modular, and scalable.</li> <li>The desirable shape, size, and crystallinity of nanoparticles can be controlled by monitoring the synthesis parameters to achieve the desired properties.</li> </ul>	<ul> <li>Chemicals potentially hazardous to human health and the environment.</li> <li>Formation of toxic byproducts.</li> </ul>	- Bottom-up approach (includes building structures from atom scale or molecular levels to the desired size.	[28- 30]				
Biological	- Involves the function of microorganisms, bacteria, fungi, algae, microalgae, yeast, and plant extracts (as reducing agents) to synthesize nanoparticles from aqueous solutions of metal salts	<ul> <li>Environment-friendly (green synthesis method)</li> <li>Cost-effective.</li> <li>Provide a convenient one-step synthetic method.</li> </ul>	<ul> <li>May contain minor toxic contaminations on the particle surface.</li> <li>Heterologous size and morphology of NPs.</li> </ul>	- Bottom-up approach.					

Synthesis method	Description	Advantages	Disadvantages	Notes	Ref.
			- Difficult extraction and isolation procedure. - Low production yield		

When compared to silicon as a standard covalent semiconductor, MOS define a class of unique materials due to their electronic charge transfer characteristics. It is a valence compound with a strong ionic bonding strength. The metal (M) ns and oxygen (O) 2p orbitals dominate their conduction band minimum (CBM) and valence band maximum (VBM), respectively. Because of the interaction between the metal and oxide orbitals, the charge carrier transport is significantly different. Generally, the orbitals of metal nanoparticles are greatly dispersed, whereas the O 2p orbitals are concentrated, resulting in a lower effective mass for electrons relative to holes. Because the carrier mobility, m\*, is inversely proportional to the carrier effective mass,  $m^*$ , via = e/m<sup>\*</sup>, where is the free carrier scattering time, the smaller the effective electron mass in metal oxide, the better the electron transport compared to hole transport. The conductivity of typical metal oxide semiconductors like In<sub>2</sub>O<sub>3</sub>, ZnO, and SnO<sub>2</sub>, as well as their solid solutions, is mostly n-type. Although these metal oxide semiconductors' electronic band topologies assist electron or whole transport, good n-type or p-type conductivity cannot be produced intrinsically. Intrinsic point defects act as donors or acceptors in metal oxide semiconductors. In many circumstances, metal oxide band gaps are often too wide, and defect levels are too deep to produce high-concentration carriers. To achieve moderate or high conductivity, extrinsic doping with a regulated concentration up to a very high level is required [31].

Photocatalytic applications could benefit from semiconductor/metal (oxide) hybrid materials. The involved mechanisms, however, are complex: the absorption properties of the light harvesters must be considered, the transfer of charges must be avoided by avoiding unwanted de-excitation pathways, and the chemical reaction must, at the very least, yield the desired product without generating unwanted side-products. Understanding the features of the potential system, albeit challenging to acquire, could be a crucial step toward the custom creation of suitable photocatalytic materials. The clarification of the underlying reaction mechanisms has greatly helped state-of-the-art systems, as seen in the total water-splitting reaction. However, the systems' complexity and utilization of multiple components have greatly expanded, but they still cannot compete with the benchmark system of water electrolysis in combination with solar cells. However, the water-splitting process is unlikely to be an optimum field of application for photocatalysis, and research has switched steadily toward carbon fixation from CO<sub>2</sub>. Furthermore, as the use of titanium for self-cleaning windows and water treatment demonstrates, semiconductor/metal (oxide) hybrid materials may have applications besides sustainable fuel production. Instead, these systems may assist in replacing high-temperature catalytic processes with photocatalytic processes and developing new synthetic routes for compounds of interest [32,33].

Wide-band-gap semiconductors like  $SnO_2$ , ZnO,  $WO_3$ , and  $TiO_2$  have a change in electrical conductivity proportional to the composition of the gas surrounding them. As a result, they are the most widely used and useful sensing materials for manufacturing low-cost gasdetecting devices. In these sensing materials,  $H_2O$  is adsorbed on the oxide surface in molecular and hydroxyl forms. The conductivity of n-type ceramics increases, whereas the conductivity of p-type ceramics decreases. This phenomenon has been explained as the transfer of electrons from the water molecules that have chemically adhered to the ceramic surface. An alternate hypothesis put out the idea that the water molecules would take the place of the previously adsorbed and ionized oxygen ( $O^-$ ,  $O^{2^-}$ , etc.), releasing the electrons from the ionized oxygen in the process. Thus, either or both of them could cause the "donor effect". The surface concentration of electrons causes conductivity, and this sort of sense is sometimes referred to as the "electronic type". On the other hand, the layer of water that has been physically absorbed might be relatively proton-conductive. We conclude that the conductivity of ceramic semiconducting materials results from the addition of electrons and protons (ionic) at room temperature as a result of this discovery. However, moisture cannot efficiently condense on the surface (mostly at the oxygen sites) and draw electrons there. Adsorbed oxygen initially causes a depletion region to occur. However, the freed electrons may be able to reverse the depletion. Adsorbed water molecules increase the conductivity of n-type ceramic semiconductors, as has already been mentioned; thus, nearly all of the research on n-type ceramics has been published too far [34,35].

Different parts of electronic devices are manufactured from varied and specific raw materials. The choice of these materials for a particular electronic component depends on its characteristics compatible with its purpose. Materials used to build up electronic devices can be located in three categories, i.e., electrotechnical, constructional, and special. Electrotechnical materials have specific electromagnetic characteristics and could be classified according to their electrical behavior into conductors, insulators, and semiconductors. While according to their magnetic behavior, they are either magnetic or weekly magnetic [6,36].

# 8. Environmental Applications of MO Semiconductors

Recent life almost depends on electric devices, mainly realized in semiconductor applications. Semiconductors can control conductivity over a wide range of temperatures and electrical and magnetic fields. Due to their miniature size, low price, and special electrical properties, these devices find applications in almost every household appliance and gadgets like gas lighter, security alarms, radios, TV, telephone, tape recorder, CD player, computers, fan regulators, emergency lights, etc. All control mechanisms in big industries, airplane flight control equipment, and satellite power systems use semiconductor devices. In a way, it isn't easy to imagine life without these [37].

Fields of communications, industry, environment, agriculture, and healthcare benefit from semiconductors devices. Semiconductor-based devices are present almost everywhere humans are today. All electrical equipment, telecommunications, computers, and sensing depend on semiconductors. Common electronic devices such as cell phones, computers, TVs, CD players, and MP3 players couldn't be works without semiconductors. The medical and industrial electronics sector has benefited greatly from the tremendous development in the field of semiconductors, which has made devices more accurate and reliable [2]. Semiconductors also play a major role in the fields of solar cells, wireless communications, spectroscopic ellipsometry, photodiode detectors, and optical communication [1].

## 8.1. MOS as Photocatalysts and environmental applications.

The environment is exposed to many organic pollutants such as pesticides, crude oil, dyes, etc., and microbial pollutants, which represent a real threat to human health and lifestyle.

Although there are several ways to get rid of these contaminants, one of the most effective and promising methods is the advanced oxidation process (AOP) [38]. Photocatalysis is an advanced oxidation process that utilizes natural or artificial UV radiation to "break down" organic matter or oxidize inorganic matter. MOS materials could solve many environmental problems, such as air and water pollution, making "self-cleaning surfaces" and building materials [12]. One of those MOS materials that are applied extensively as photocatalysts are TiO<sub>2</sub>. Titanium oxide is widely applied to oxidize and degrade several types of volatile and non-volatile organic pollutants in a gaseous or liquid state. Kiriakidis and Binas [39] recorded successful experiments for textile dye degradation using "Mn-, Co and Mn/Co-doped TiO<sub>2</sub> semiconductor powder materials". The co-precipitation synthesized nanomaterial of Mn-, Co, and Mn/Co-doped TiO<sub>2</sub> were significantly efficient under visible light for wastewater purification. In addition to organic pollutants, heavy metals could also be removed from wastewater by MOS as "small crystallites deposited on the surface of the photocatalyst according to the redox process". Additionally, valuable metals like platinum, gold, and silver can be recovered from industrial effluent using photocatalysis's photoreduction capability [40].

#### 8.2. MOS in energy.

Metal Oxide semiconductors have many applications in the field of energy production and conversion. In solar cells, most binary metal chalcogenides, such as PbS, PbSe, CdS, CdSe, and  $Ag_2S$ , are usually utilized due to their capacity to adsorb within the band gap of 1.1 to 1.4 eV. However, light harvesting and energy conversion efficiency are quite low, prompting a material modification to increase efficiency. This modification can be made by combining semiconductors with different bandgap materials such as CdS/ZnO, PbSe/TiO<sub>2</sub>, etc. In addition, MOS materials/nanomaterials are widely used to replace Si in solar due to their low cost, tunable band gap, multiple electron-hole pair generation, optical absorption coefficient, and quantum size effect. In particular, thin-film photovoltaic cells are lightweight and flexible and can fit a variety of surfaces to generate power from sunlight or ambient light [12,41]. One of the most researched and often used nanomaterials from the layered transition-metal dichalcogenides (TMDs) semiconductor family is the molybdenum disulfide (MoS<sub>2</sub>). This is because MoS<sub>2</sub> has the characteristic of "large carrier diffusion length" and "high carrier mobility" [42-43]. N-type silicon (n-Si) together with single-wall carbon nanotube (SWCNT) and MoS2 has been applied to manufacture "novel SWCNT/n-Si photovoltaic devices" by Almalki et al. [43]. They covered the Si structure of the solar cell with a "MoS<sub>2</sub> thin film" which was subsequently covered by an "SWCNT film". The photoconversion efficiency (PCE) of the SWCNT/n-Si solar cells has doubled as a result of this MoS<sub>2</sub> hybrid insertion.

Hydrogen production by water splitting (into  $O_2$  and  $H_2$  gases) is an environmentally friendly process to replace a portion of fossil fuel with a sustainable green source. This process could be performed by the activation of "suitable photocatalysts" using solar energy. The characteristic of the "suitable photocatalysts" for energy generation includes its chemical stability, corrosion resistance, and radiation absorption ability within the visible region. Many "submerged" metal oxides have the ability to "absorb radiation in the visible region", have low bandgap energy, and therefore have the advantage of being used as an efficient photoelectric hydrogen generator [12]. The conventional photoelectrochemical (PEC) cell consists of a photoanode and a photocathode. In the photoanode, electrons are extracted from water using solar irradiation as the energy source, and in the photocathode, reductive electrons are used for the hydrogen-generation reaction [44]. Figure (6) represents a photoelectrochemical water splitting system using an n-type semiconductor photoanode.



Figure 6. Photoelectrochemical water splitting systems using an n-type semiconductor photoanode.

In addition to naturally generated pollutants, human activities cause an increase in many types of pollutants that are dangerous to the environment and humans themselves. These pollutants must be constantly monitored and reduced to preserve humans and the environment [12]. On the other hand, sensors are of great medical importance, as many vital factors must be measured and followed up to diagnose diseases and thus determine appropriate treatment modalities. Due to the important requirements in detecting and measuring many pollutants and biological agents, scientists have developed many related sensors in many fields. Conceptually, the mechanisms governing sensing reactions of different parameters (e.g., gas, humidity, and UV) are identical. The response mechanism in these sensors mainly depends on the change in electrical conductivity or resistance of metallic oxide semiconducting materials when exposed to different materials [45]. Sensors could be designed to detect or measure specific element(s) in the surrounding environment (soil, water, and air) by controlling the detector material(s). MOS material, particularly n-type semiconductors, plays an important role in sensing selectivity.

For instance, ZnO, PbSe, SnO<sub>2</sub>, WO<sub>3</sub>, and PbS colloidal quantum dots are used to improve the selectivity of gases sensors towards some gases such as NH<sub>3</sub>, NO<sub>2</sub>, LPG, CH<sub>4</sub>, ethanol, and H<sub>2</sub>S. At the same time, SnO<sub>2</sub> is applied as doping material to increase the selectivity of Nanogab sensors towards alcohol metabolites (detect drunk driving). Moreover, reduced graphene oxide (rGO) incorporated with SnO<sub>2</sub> is widely used for selective heavy metal detection in drinking water [12].

## 8.3. MOS in sensing devices

## 8.3.1. Sensors and biosensors.

There is a need for rapid and accurate monitoring of air and water quality and for detecting and estimating specific elements in various environmental, industrial, and biological samples. However, the conventional detection and quantification methods for the elements and pollutants are relatively expensive, time-consuming, and depend on highly trained persons. Moreover, usually, samples need to be transferred to laboratories for analysis purposes which

could influence the analysis quality and cost. Therefore, the need has arisen to develop rapid, accurate, available on-site, and cost-effective unconventional instruments and these advantages can be realized through biosensors and sensors [46-48]. A sensor is a device or instrument that can detect and respond to a parameter or "analyte" and refer to it in an understandable form. The sensor typically consists of a "receptor" that responds to the "analyte" and transfers this response to the "transducer" and "detector" that outputs this response via a "read-out unit" [49,50]. The analyte could be a physical parameter such as temperature, humidity, blood pressure, etc., or an element in gas or liquid. Biosensors are "combined and integrated analytical tools based on specific interaction(s) between the target analyte and a specific builtin bio-receptor, in which the response changes in the biomolecule converted proportionally into measurable signals by the transducer". The bioreceptor in biosensors could be living cells (microbial, plant, or animal cells) or some of their bio-components (enzymes, antibodies, nucleic acids, etc.) [48].

Since semiconductors are important materials in the current technology, they can play an important role (as electronic transducers) in sensors and biosensor construction. Semiconductors are employed to use the potential charge differences in the electric field sensitively. The doping material plays an important role in detection and sensing, where it is affected by the biological elements, thus changing the potential difference across the sensorsensitive material and "control the current between source and drain of transistors junction" [51].

#### 8.3.2. Sensing based on metal oxides.

Metal oxide semiconductors have great potential to develop into competitive biosensor materials. This is based on their adaptability in terms of morphology [52], physicochemical interfacial properties, capability to combine in composite structures [53], and chemical stability [54]. Due to their electrochemical sensitivity, attention is gained, and they are suitable for enzyme-based biosensors due to their energy band alignment. Another key advantage is their cost-effective synthesis. Such ZnO has a stronger ability to bond with biological recognition components than other metal oxides (TiO<sub>2</sub>). This gives it a significant benefit for biomedical applications. It can be used for wearable sensors because it is non-toxic and compatible with human skin [55]. ZnO nanostars have been made via chemical bath deposition (CBD) and are being utilized to identify microRNA in cancer cells. Prior to the creation of ECL biosensors, the surface was functionalized utilizing thiol-modified hairpin and hybridization chain processes. This substance is a strong candidate for a clinical bioassay because the LOD was determined to be 18.6 pM. The same method was created to identify the Zika virus in undiluted urine [56-57]. The Zika virus causes symptoms including headaches, arthralgia, myalgia, or conjunctivitis and is spread by mosquito bites. The ZIKV-NS1 antibody was immobilized on the ZnO nanoparticles using glutaraldehyde and cystamine. The outcome of the CV evaluation of the LOD was 1.00 pg/mL [56].

The hydrothermal process is another method that has been widely employed for ZnO production with bio-sensing applications. Hydrothermally produced ZnO nanoparticles and nanorods were both used in biosensors to detect glucose [58,59]. The limit of detection using differential pulse voltammetry for the functionalized ZnO nanorods with glucose oxidase was 1.0 M. When compared to ZnO nanopowder functionalized with glucose oxidase. Their results are noticeably improved, detecting as low as 50 M. Hydrothermally produced ZnO nanorods were employed as phosphate and IgG detection sensors [60]. ZnO was functionalized with https://biointerfaceresearch.com/

pyruvate oxidase by immersion for phosphate detection, and the LOD was 0.5 M. The ZnO surface was functionalized with myoglobin by immersion and cold drying in order to create a G immunoglobulin sensor with a 0.03 ng/mL LOD. The hydrothermal method was used to create ZnO nanocone arrays for dopamine detection [61], and ZnO electrodes were employed to detect serotonin [60-61].

Additionally, metal oxide nanoparticles have distinct optical qualities that depend on surface chemistry. As an illustration, nanoceria dispersion is nearly colorless, but when  $H_2O_2$ is added, it immediately turns yellow and dark orange. The rise in surface Ce (IV) and surface peroxide species was blamed for the color shift. In this colorimetric test, the linear range for H<sub>2</sub>O<sub>2</sub> detection was 0.01 to 0.15 mM. When paired with glucose oxidase, this could then be used in a paper-based test and allowed for the detection of glucose [62].

Advancement resulted from the long-known ability of magnetic iron oxide nanoparticles to absorb aqueous arsenic species. It is challenging to investigate the process, though. A quantitative fluorescent assay was created due to integrating Fe<sub>3</sub>O<sub>4</sub> nanoparticles' capacity for fluorescence quenching and DNA adsorption [63]. The assay's foundation was the competitive adsorption of As(V) and DNA modified with 6-carboxyfluorescein (FAM) on the surface of Fe<sub>3</sub>O<sub>4</sub> (Fig. 7). As(V) could be detected by this fluorescence turn-on sensor at concentrations as low as 300 nM (or 130 nM of As(V) was pre-adsorbed to prevent DNA adsorption). Except for phosphate, which is extremely similar to arsenate, this displacement test was selective for only a few significant anions.



Figure 7. Adsorption of arsenate by iron oxide.

#### 8.4. Metal oxide semiconductors as the gas sensing material.

Due to the ability to control their physical and chemical properties, metal oxides are the foundation for modern intellectual and functional materials and technologies. Numerous chemical and structural factors, such as chemical composition, structural flaws, morphology, grain size, specific surface area, and others, affect the functional qualities of MOxs. Any one of these traits can be altered to control the properties they possess. As a result, MOxs are the most diversified family of materials with properties that cover practically all areas of materials science and physics in the domains of semiconducting, superconductivity, ferroelectricity, and magnetism, thanks to their distinctive qualities [64]. For semiconductor gas sensors, sensing materials such as SnO<sub>2</sub>, ZnO, In<sub>2</sub>O<sub>3</sub>, tungsten trioxide (WO<sub>3</sub>), CdO, TiO<sub>2</sub>, and other MOxs can be employed. Due to a special set of functional characteristics, the most significant of which are electrical conductivity, transparency in a broad range of spectra, and strong surface https://biointerfaceresearch.com/

reactivity, they are classified into a group of transparent conductive oxides.  $SnO_2$  and  $TiO_2$  are the most widely used SMOxs. Nevertheless, because of their ease of manufacture, affordability, great chemical stability, mechanical strength, heat resistance, and high adherence to glass and other substrates

Sensitivity, response and recovery time, selectivity, stability, detection limit and resolution, and working temperature are the most crucial quality indicators of gas sensor performance. By reducing, the particle size to the nanoscale, doping (modifying) the sensing material, and enhancing sensor design, these properties of gas sensors based on SMOxs can be greatly enhanced. The porosity of the sensitive material, the working temperature, the presence of dopants or modifiers, and the crystallite size substantially impact the sensitivity of gas sensors [65,66]. Controlling the particle size of semiconductor materials is one of the first needs to increase the sensor's sensitivity because sensing reactions primarily occur on the surface of the sensitive material. Due to their high specific surface area and hence increased adsorption capacity, nanocrystalline materials are known for having the highest sensor signal values [66]. It has proven able to detect H<sub>2</sub> at a sub-ppm level by lowering the particle size of the sensing material [67]. Moreover, it was demonstrated that at lower temperatures, the sensing materials based on TiO<sub>2</sub> NPs offer strong selectivity and sensitivity toward H<sub>2</sub>S, CH<sub>3</sub>OH, and C<sub>2</sub>H<sub>5</sub>OH gases and possibilities for humidity detection even at room temperature. Thus, sensors with high mechanical stability, quick reaction and recovery times, strong reproducibility, low energy consumption, and small size and weight were developed [68,69].

Selectivity refers to the semiconductor layer's capacity to identify a particular target gas or group of target gases in a gas mixture. Surface modification or bulk doping with different catalytic additions is used to improve the target components' ability to adsorb in order to boost the selectivity of gas sensors [70,71]. Previous research shows that  $SnO_2$  and  $TiO_2$ nanostructure-based sensing materials produce high selectivity sensors through either surface NM loading or bulk doping with redox-capable elements, enabling selective gas detection in mixed gas settings. These findings show that developing new active centers on the MOx surface or altering the material's electrical structure is responsible for improving sensing performance in such circumstances [67,72].

Response time establishes the time frame in which the parameter value at a certain gas concentration changes by a specific percentage from its original value. Through the doping of MOxs with NMs, the response time can be slashed. Choi et al. [73] showed that Pd-doped SnO2 sensing material exhibits a reaction time of fewer than 10 s in this manner. The response time of 0.08 weight percent Pt-doped  $SnO_2$  nanofibers to  $H_2S$  is substantially faster (1 s) than the response time of the undoped SnO<sub>2</sub> nanofibers (2-7 s), according to Dong et al. [74]. The recovery time is the time needed for the sensor to return to its initial value after the target gas concentration is reduced to zero. The slow surface reactions that cause the prolonged recovery time can be sped up by doping metal catalysts like Pd and Ag. When exposed to H<sub>2</sub>, CO, CH<sub>4</sub>, and C<sub>2</sub>H<sub>5</sub>OH, Pd-doped (0.4 wt%) SnO<sub>2</sub> hollow nanofibers show a dramatically shorter recovery time (31.8, 23.7, 38.5, and 88.5 s, respectively). In this instance, the Pd catalytic enhancement of the oxygen adsorption can account for the longer recovery time [72]. The operating temperature affects response and recovery times. The sensitivity of TiO<sub>2</sub> nanofibers to NO<sub>2</sub> declines with temperature rise, whereas response time lowers with temperature growth and decreases with growth in NO<sub>2</sub> concentration, according to research by Landau et al. [75] on the effect of temperature on sensing properties.

The lowest gas concentration that a sensor element can detect known as the detection limit. The resolution of the instrument is determined by the smallest concentration difference that the sensor can detect. The temperature at which a sensor operates at its highest level of sensitivity is known as the working temperature. In addition to the properties of the sensitive material, the sensor's schematics, design, and dimensions are crucial in enhancing sensor performance [76]. According to the study by Souhir *et al.* [77] on three distinct metals (platinum, titanium, and tungsten) used in heater manufacture, platinum has the best prospects since it offers a good balance between high temperature and power consumption. The power consumption can be decreased by reducing the thickness of the heating electrode and the intertrack in the heater's design, which improves temperature homogeneity. The electrodes, specifically the electrode geometry and gap size, are the other structural component of the MOx sensing element that significantly impact the gas sensing characteristics.

## 8.5. The mechanism of N-type MOx for gas sensing.

Fundamentally, the MOxs' gas sensing mechanism is a surface-related problem that causes a change in sensor resistance that is closely tied to the target gases' adsorption or desorption processes. Romppainen, first of 1988, suggested the potential operating principles of the n-type SMOx-based sensing mechanism. He asserts that the following processes will have an impact on the surface conductivity, which he claims is the primary determining element for the sensing mechanism:

- Oxidation/reduction of the semiconductor: Sensitive materials' charge transport concentration and electron structure are impacted by surface or bulk stoichiometry and gas content changes.

- Ion exchange: In contrast to the base material, the change in ions causes the surface layer to develop with a different electron structure.

- Gas adsorption: The gas molecule adheres to the material's surface and either contributes electrons to the conducting band or serves as a hub for trapping particles.

- Reaction with adsorbed species: The surface's amount of trapped electrons is reduced as a result of the adsorbed gas species' reaction with previously deposited oxygen.

Oxygen molecules are adsorbed on the surface of the n-type nanostructure after exposure to air. The establishment of potential barriers on the particle surface is caused by the transfer of electrons from the semiconductor powder to the surface. This results in a comparatively small number of oxygen adsorption sites on the n-type semiconductor surface. To go from one particle to the next for conductance, electrons must cross this surface barrier [78]. Adsorbed oxygen ion species ( $O^{-2}$ ,  $O^-$ , and  $O^{2-}$ ) are created by the extraction of conduction band electrons and creating a space charge conduction region, depending on the temperature. The sensor resistance rises as a result. As soon as the target gas is injected, it is adsorbed on the material's surface. If it is a reducing gas, it undergoes oxidation, which causes electrons to return to the conduction band. This has the effect of decreasing the space charge width and the resistance proportional to the analyte concentration. When gas oxidizes, it reduces by removing electrons from the conduction band, which causes the resistance to rise proportionately [79,80]. Thus, Reducing gases increase the conductivity of n-type semiconductors, while oxidizing gases reduce the sensor's conductivity.

#### 8.6. N-Type Metal Oxide Semiconductor Materials for gas detection: case study

Air pollution is a real concern because it affects human health and lifestyle. Therefore it is necessary to disclose it to reduce its negative impact on the environment and human beings. On the other hand, for safety purposes, the leakage of flammable or toxic gases must be detected and linked to a control system to enable automatic shutdown. The air pollutants are located under three major groups, i.e., inorganic gases, organic gases, and particulate matter. Inorganic gases include carbon, sulfur, and nitrogen oxides as well as H<sub>2</sub>S, NH<sub>3</sub>, C<sub>12</sub>, etc.). Organic gases include "CH<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, C<sub>3</sub>H<sub>8</sub>, C<sub>2</sub>H<sub>2</sub>, C<sub>2</sub>H<sub>4</sub>, C<sub>8</sub>H<sub>18</sub>, C<sub>6</sub>H<sub>6</sub>, formaldehyde, the vapor of acetone, alcohol, organic acid, petrol, diesel, LPG containing volatile organic compounds". Particulate matters that could pollute the air could be "dust, smoke, ash, carbon, lead, sprays, insecticides, pesticides, oil, grease, paint, etc." [81].

Many gas sensors are made based on MOS; the sensing layer helps to determine the selectivity and sensitivity of the sensor [82]. The gas sensing process is highly dependent on the surface interactions on the sensing layer. Gas adsorption or adsorption on materials mounted on thick/thin films of MOS causes a significant change in the material's electrical resistance (conductivity). The measurement of changes in capacity, work function, mass, optical properties, or reaction energy released by a gas/solid interaction is also used to detect such interactions in a gas-sensing material [81]. As mentioned earlier, based on the type of charge carriers, MOSs are classified into n-type (e.g., ZnO, SnO<sub>2</sub>, WO<sub>3</sub>, etc.) and p-type (e.g., NiO, CuO, O<sub>3</sub>, and CO<sub>2</sub>). Although semiconductor gas sensors have many advantages, such as high sensitivity, small size, promptness, and low cost; however, the main drawback of these sensors may be poor selectivity. Therefore, choosing the electron donors or acceptors (impurity type) of the sensor layer helps improve the sensor's selectivity and sensitivity [82,83]. Figure (8) represents a general composition for gas sensors.



Figure 8. General composition for gas sensors.

In a study on sensing H<sub>2</sub> and CO (reducing gases), Yin *et al.*, [84] stated that MOSbased sensors "cannot detect the concentrations of both gases because the MOS sensors have cross-sensitive behavior to H<sub>2</sub> and CO". Hence, they prepared a gas sensor based on SnO<sup>2-</sup> Mn<sub>3</sub>O<sub>4</sub> nanocomposites that "show n-type response to H<sub>2</sub> and CO for the molar ratio of Sn to Mn greater than 1:2.33, whereas the composites show p-type response to H<sub>2</sub> and CO for the molar ratio of Sn to Mn smaller than 1:2.33". When the ratio of Sn to Mn is adjusted to be 1:2.33, the sensor behaves as n-type for CO while it behaves as p-type for H<sub>2</sub>. This indicates the capability to increase the sensor selectivity based on the modulation of "current carrier materials".

For detecting highly explosive gases such as methane, MOS gas sensors could interfere with "a heavy cross-sensitive response to volatile organic compounds such as ethanol and acetic acid". Wu *et al.* [85] concluded that filtration could "help" the MOS sensor to be more selective for methane. They designed an "on-chip microfilter" fabricated from a porous alumina ceramic and loaded it on the surface with platinum. They found that the detection selectivity and sensitivity to methane were enhanced by the MOS (tin dioxide and indium oxide) sensor due to the application of "the on-chip microfilter".

For nitrogen dioxide sensing, Tyagi *et al.* [86] examined the performance of two thinfilm MOS sensors using pure MoS<sub>2</sub> or Cu-MoS<sub>2</sub>. Under the study conditions, i.e., 100 °C and 2-200 ppm of NO<sub>2</sub>, the Cu-MoS<sub>2</sub>-based sensor presented the highest and fastest sensing response at 20 ppm of NO<sub>2</sub>. The same study concluded that "the Cu-doped MoS<sub>2</sub> sensor can detect a trace amount of NO<sub>2</sub> down to 2 ppm at a low operating temperature of 100°C".

Volatile organic compound (VOCs) vapor is an important parameter in many industries. It's sensing using MOS-based sensors could be affected by air humidity. Hence, Yan *et al.* [84] examined four different MOS sensors "in response to VOCs at different humid environments". They concluded that the water vapor in the air interferes with the response and sensitivity of MOS sensors. Munusami *et al.*, [87] designed (and fabricated) a hybrid gas sensor based on MoS<sub>2</sub>/graphene to detect various types of gases (LPG, ethanol, methanol, acetone, CO, and NO<sub>2</sub>). The sheet of MoS<sub>2</sub> spherical nanoparticles and 2D graphene mixture provides a large active surface area, "making it possible for gas molecules with evanescent wave light to absorb more properties". The designed hybrid sensor showed high sensitivity (71%) and fast response (15 s) for LPG gas detection. In addition, it exhibited high stability as it could retain 96% of its original sensitivity and low recovery time (22 s).

Particularly at high temperatures and challenging situations, semiconductor metal oxides are thought to have great promise for hydrogen sensor applications [88]. Metal oxide thin films and nanostructures were used to create a thorough assessment of hydrogen gas sensors [89]. It is thought that the reduction in resistance caused by the interactions of chemisorbed oxygen species with hydrogen is what causes metal oxide to sense hydrogen [90]. Ga<sub>2</sub>O<sub>3</sub> thin films may be used to detect reducing gases at high temperatures. In particular, the first demonstration of a hydrogen sensor utilizing a Pt Schottky diode on a thin sheet of Ga<sub>2</sub>O<sub>3</sub> [91]. The researchers demonstrated that increasing hydrogen concentrations effectively regulated the Schottky barrier height of Pt/Ga<sub>2</sub>O<sub>3</sub> diode sensors. A field-effect hydrogen sensor with improved response to hydrogen and stable operation at high temperatures beyond 400°C was described employing Ga<sub>2</sub>O<sub>3</sub> thin film and Schottky diodes based on  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> single crystals. As stated in this section, 2-01 and (010)  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> single crystals have been used to examine the hydrogen sensing properties of Pt Schottky diodes [92].

For the device's construction, Ti/Al was used to create Ohmic contacts on  $\beta$ -Ga<sub>2</sub>O<sub>3</sub>, which was then deposited by e-beam evaporation, patterned by lift-off, and then quickly heated in a setup with a nitrogen atmosphere. Using plasma-enhanced chemical vapor deposition, a SiNx passivation layer with a 200 nm thickness was created for diode isolation. Buffered oxide etching was used to create the windows for the active sensing area. Ti/Au contact pads were applied to the Schottky contact area after the 10-nm-thick Pt coating to allow for wire bonding and probing. An Agilent 4155C semiconductor parameter analyzer was used to examine the I-V characteristics of the Pt Schottky diode sensors on 2-01 and (010)  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> under the

flammable limit of hydrogen (4 %) balanced with nitrogen in a gas test chamber [92]. Figure (9) summarizes the applications of MOS.



Figure 9. A general summary of MOS applications.

# 9. Conclusions

Semiconductors, especially N-type metal oxide semiconductors (MOS), are particularly interesting due to their unique properties and wide applications. Their importance covered almost all fields of life, including electronic devices, environmental applications, solar cells, hydrogen cells, and sensor applications. Incorporating nanoparticles as doping materials for semiconductors could enhance their functions as photocatalysts in fuel cells, solar cells, and environmental applications, as well as selectivity and sensitivity in the case of sensors.

# Funding

This research received no external funding.

# Acknowledgments

The authors would like to thank the National Research Centre (NRC), Egypt, and the College of Medicine, Al-Qunfudah Branch, Umm Al-Qura University, Makkah, Saudi Arabia, for providing the necessary facilities to carry out the research work.

# **Conflicts of Interest**

The authors declare no conflict of interest.

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