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Carbon Nano Based Materials and Their Composites for Gas Sensing Applications: Review

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Carbon Nano Based Materials and Their Composites for Gas Sensing Applications: Review

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Abstract

The consequences of carbon nano materials incorporation for gas sensing applications received noteworthy consideration in the recent preceding years. Owing to their desirable physical and chemical specification, the carbon nanomaterials and combinations with other materials are well suited for the gas sensing applications as it satisfies the condition of elevated surface area and reduced bandgap. Herein, we start this review by reviewing the importance and preparation methods of carbon nanomaterials. Varied carbon based nanomaterials as carbon nanoonions, carbon nanohorns, nanodiamonds and carbon quantum dots are explored. Graphene and its derivatives are normally combined with CNTs, thus. We introduce after that the graphene synthesis, specifications, and graphene/metal oxides, noble metals, and polymers nanocomposites for gas sensing. This review summarized the keypoints for the gas sensing area that relied on carbon nanomaterials. Furthermore different types of sensors toward different gas species have been reported and discussed.

Keywords:Graphene; CNTs;Gas sensors; Nanomaterials; Carbon nanohorns

Outline

1. Introduction

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Carbon is one of the most abundant elements on the planet, as it grasps the ability to produce anenormous number of bonds that can be formed with different materials or with itself. Besides, carbon allotropesexistsas the carbon can hybridize in sp, sp2 and sp3 configurations.

From another point of view, carbon is exclusive among the elements due to its ability to adopt extended two-dimensional sheet structures in elemental form. Since the in-sheet bonding is tremendously strong, these sheets are stable both as isolated objects (graphene), and when turned into cylindrical geometries (nanotubes) or quasispherical geometries (fullerenes) wherein, e.g., pentagonal rings provide the needed Gaussian curvature. The carbon nanocone, is an intermediate state between graphene and fullerene wherein a single pentagonal ring or assembly of nearby pentagonal rings defines a conical apex, which is then extended by a pure-hexagon graphenic network into a larger conical structure [1].

In this review,we shed light on the carbon nanomaterials and their composites with different materials (i.e., metal oxides, polymer, metals, etc.) and theirapplications as gas sensors. These carbon nanomaterials comprise carbon nanohorns, carbon nano-onions (CNOs), nano diamond, carbon quantum dots (CQDs), carbon nanotubes (CNTs), and Grapheneas represented in fig. 1.Remarkable attention has been devoted to bothCNTs and graphene as they are extensivelyconsidered. In what follows we point out the most characteristic sorts of carbon nanomaterials.

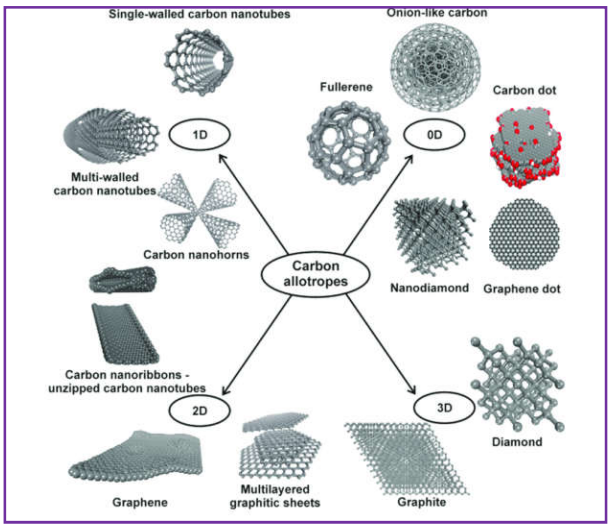

Figure 1: The illustration of allotropes of carbon nanomaterials, from [2]CC BY 4.0

2. Carbon nanohorns (CNHs):

Carbon nanohorns (CNHs) is belonging to the carbon nanomaterials family that have horn shaped tips and are closely similar to single wall carbon nanotubes (SWCNTs) as exposed in fig. 2. The essential characteristics of CHNs are their affinity to group together to form spherical clusters with overall dimensions of 2–5 nm in diameter and a length around 40–50 nm [1]. The three different forms of CNHs are dahlia-like flowers, bud-like flowers, and seed-like flowers [3]. The despite their unique advantages that are represented as high surface area, elevated electrical and thermal conductivity, and ease of functionalization, they are synthesized by laser ablation of pure graphite without the use of catalyst at room temperature with high yield and production rate[4][5].

Due to their high surface area and a large number of cavities, CNHs are promising candidates in different applications [6]like drug delivery [7], magnetic resonance analysis [8], photodynamic therapy [9] biosensing [10] and gas sensors . Suehiro et al. utilized dielectrophoresis (DEF) technique to fabricate CNHs gas sensor. The DEP-fabricated CNHs sensor was evaluated toward nitrogen dioxide (NO2) and ammonia (NH3) gas utilizing impedance spectroscopy. They demonstrated that the CNHs could detect the presence of ppm–levels of NH3 and NO2 gases at room temperature [11]. Sano et al. reported a simple and cost-effective method for producing CNHs and studied their gas sensing properties. They confirmed that the CNHs gas sensor can detect NH3 and O3 at room temperature. Their results demonstrated that gas sensors based on CNHs exhibited higher sensitivity compared to the sensor based on SWCNTs at the same conditions. They correlated this enhancement to mono-layer gas adsorption and the interaction of adsorbed gas molecules which affects the charge transfer from gas molecules to the sensor surface [12]. The applicability of carbon nanohorns can be extended as well for humidity sensing with the oxidized nanohorns as an active sensing layer. sensing performance was tested within a 10-90% humidity of nitrogen and air. The sensitivity in air outcomes the sensitivity in humid nitrogen with a factor of two. carboxylic groups existence can clarify this as the dealings with water molecules (as electron donors) declines the number of holes and oxidized single-walled carbon nanohorns develops higher resistivity [13]

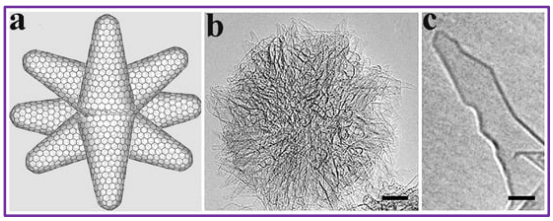

Figure 2:(a) Schematic demonstration of the carbon nanohorns, (b) The high-resolution transmission electron microscope (HRTEM) for the nanohorns, and (c) HRTEM for separate carbon nanohorn where the scale bar is set as 2 nm, from [14] CC BY 4.0

3. Carbon nano-onions (CNOs):

Carbon nano onions (CNOs) depicted in fig 3 are an allotrope of carbon nanomaterials belonging to the fullerene family, they are made of spherical, quasi-spherical, and polyhedral with concentric graphitic shells [15][16]. CNOs consist of multiple graphitic shell structures which can be defined by the series: C60@C240@C540@C960@C1500@C60n …, where n is the shell number [17]. Many synthesizing methods like thermal annealing of nanodiamonds (NDs) [18], arc discharge [19], flame assisted pyrolysis [20], chemical vapor deposition (CVD), and non-thermal plasma [21] have been explored to obtain CNOs. The outstanding properties of CNOs represented in high specific surface area, amazing electrical conductivity, and excellent tribological behavior could be due to their exclusive shell-shaped structure [16] . The above-mentioned properties of CNOs nominate it as a suitable material to be applied in many applications such as optical limiting, catalysis, energy storage, supercapacitors, lubricants, electrochemical sensors, and gas sensors [15]. To the best of our knowledge, limited studies have been focused on the gas sensing properties of CNOs. Dhonge et al investigated the gas sensing mechanism of CNOs toward volatile organic compounds (VOCs) at room temperature. They demonstrated that there was a linear relationship between sensitivity and gas concentration in the range of 34–148 ppm [22] as displayed in fig.4.

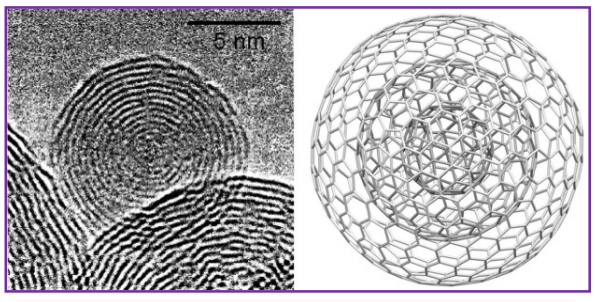

Figure 3: Carbon nanoonions, taken from[23] CC By 4.0.

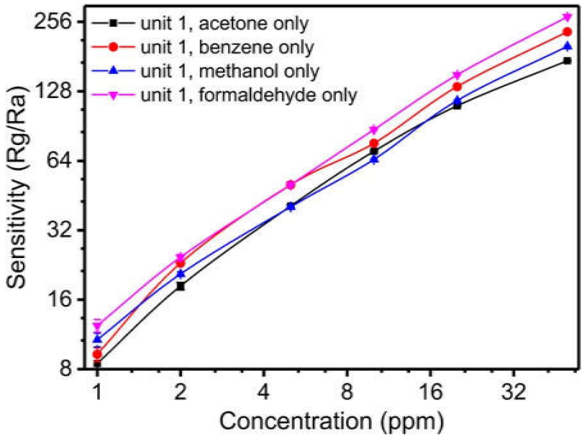

Figure 4: The sensitivity and gas concentration relationship for varied gases, obtained from [24]CC BY 4.0.

4. Nano diamond (ND):

Nano diamond (ND) is a zero dimension carbon Nano allotrope consisting primarily of short-range order tetrahedral SP3 bonded carbon atoms[25][26]. ND can be produced by three main industrial production technologies. The first technology is the mechanical grinding of high-quality diamond microcrystals produced under extreme conditions (High -temperature and high-pressure HTHP) from graphite.The second one is comprising the detonation synthesis from explosives. While the third one is CVD. The ND produced at extreme conditions is available with a particle size around 10–25 nm. The typical size of ND produced by the detonation method as estimated from XRD is 4–5nm [27]. Recently atmospheric pressure microplasma (AMP) has been considered as better choice as compared to the most commonly used detonation, HTHP and CVD [28]. NDs can be implemented in many applications such as magnetic resonance imaging, chromatography, electrochemical double-layer capacitors and batteries. Furthermore, lubrication, drug delivery and gas sensors [26][29]. This could be attributed to their exceptional chemical mechanical and optical properties. Referring to the gas sensing area of application, the synthesis of MWCNTs was executed employing nanocrystalline diamonds and the CVD procedure. In brief, the synthesized materials possess a rapid response for H2 gas detection. Further selectivity and repeatability were affirmed for a sustained duration of 2 months. The elevated response/recovery attitude might be referred to the rise of defect locations because of ND grains existence which subsequently promotes the formation of multiple binding sites for the hydrogen molecules[30].

5. Carbon Quantum Dots (CQDs):

Carbon quantum dots (CQDs) are a novel member of zero-dimensional carbon-based nanomaterials having a particle size less than 10nm in diameter whichis characterized by relatively strong fluorescence, biocompatibility, high thermal stability, inactive chemical structure and ease of functionalization [31]. CQDSs maintain a significant sum of elevated specifications, to recite some, decent photoluminance, facile preparation methodologies, low- cost, reduced level of toxicity, as well as simple functionalization. Recently, CQDs deemed the application in the sensing area with a fine detection limit within nano-, pico- or even femtomolar amounts[32]. The incorporation of ZnO onto CQDs matrix to form a composite through the conventional hydrothermal route is conducted. This composite is applied for the sensing and monitoring of nitric oxide (NO) gas. The seizing of a recovery/ response times of 34, 36 s correspondingly is attained for a 100 ppm concentration detection limitThe NO2detection is further detected using the silica aerogels functionalized CQDs. In this work, the verified selectivity of the NO2gas amongst diverse gases as O2CO2, and NH3 is confirmed [33]. Aligning with this gas sensing for carbon dots, the composite of carbon dots reinforced with MgO nanoparticles was synthesized and exploited for the detection of H2S gas using engineered Schottky apparatus. The I-V tendency for the sensor is assessed in air and targeted gas as well.

Inherently, the external voltage of -0.7 V for the 120 ppm gas concentration reports an elevated response of 11 times larger than the MgO. The noticeable diminish of the barrier height emerged during the H2S gas exposure is responsible for the raised response of the aforementioned gas than other gases[34]

Ammonia gas sensor is employed using graphene quantum dots fabricated by a simple solution fabrication method at ambient conditions. Interestingly, a flexible modulation of the pH value from acidic to neutral results in a two contrary current responses for the gas[35].

6. Carbon nanotubes (CNTs)

Throughout the preceding three decades, the employment of CNTs in diverse applications is increased due to their eminent physical and chemical characteristics. Furthermore, CNTs have been in the eyes of researchers due to their exceptional thermal, electrical, chemical properties, stability, virtuous elastic modulus (1 ~TPa), high aspect ratio (50-500), strength ~100GPa), high stiffness and low density (~1.2 g/cm3 to ~1.8 g/cm3).

Starting from 2013 the researchers directed their interest for the designing of metal-organic frameworks reinforced CNTs for the energy appliances[36][37]. Besides, they are employed in the solar system nanofluids as CNTs are significantly characterized by elevating thermal and optoelectronic specifications of the nanofluid[38]The evolving of medical applications for CNTs is provided as well thanks to the increased surface atoms ratio which might feasibly modulate the physicochemical specificationsand promote higher chemical reactivity [39]. Commonly, there are two sorts of CNTs relied on their morphologies, either as single-wall carbon nanotube (SWCNTs) or multiwall carbon nanotube (MWCNTs). TheSWCNTs are made up ofa single graphene layer rolled in a cylindrical form (Fig:5(a)) while, for MWCNTsmultiple layers of graphene (Fig:5(b)) are twisted into a cylindrical tube shape[40–42].The CNTs (SWCNTs and MWCNTs) have been examined and tested to be utilized in various applications. Nowadays CNTs can be found approximately in all applications and industries, including medicine, pharmacy, agriculture, materials science, engineering, electronics, water treatment, solar cells, etc.

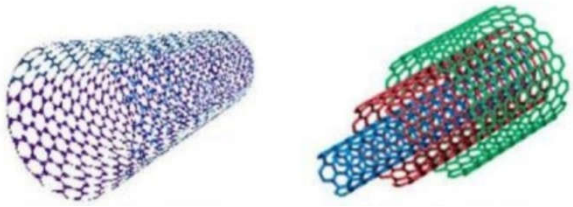

Figure 5: The visual illustration of (left) single wall carbon nanotubes (SWCNTs), and (right) multiwall carbon nanotubes (MWCNTs), acquired from [23]CC BY 4.0

6.1. CNTs synthesizing techniques:

Due to their outstanding properties, many synthesizing methods have been utilized for CNTs. All of these methods are categorized into three main techniques, stated as: arc discharge[43], laser ablation[44], and CVD[45] as represented in fig 6. The first utilized technique was arc discharge which consists of two carbon electrodes (anode and cathode) connected to low voltage high current DC power supply underinert media.The anode is filled with a mixture of powdered carbon precursorsand catalysts.

The arc discharge is the first introduced technique for CNTs synthesizing.Simply described as the generation of plasma because of the electrical breakdown of the gas[46]. Generally, an arc is

introduced between a couple of graphitic rods separated by few nanometers. This process produces highly pure and defect-free CNTs, and it delivers a low cost and reduced time compared with other methods for CNTs synthesis[47].

However it suffers from many drawbacks represented in low yield which suppresses its practical applications[46, 48]

The laser ablation method for CNTs synthesizing comprises the evaporation of carbon from high pure carbon electrode using a laser beam at elevated temperatures. The laser ablation method is not extensively utilized as it consumes high energy and requires expensive equipment [43].

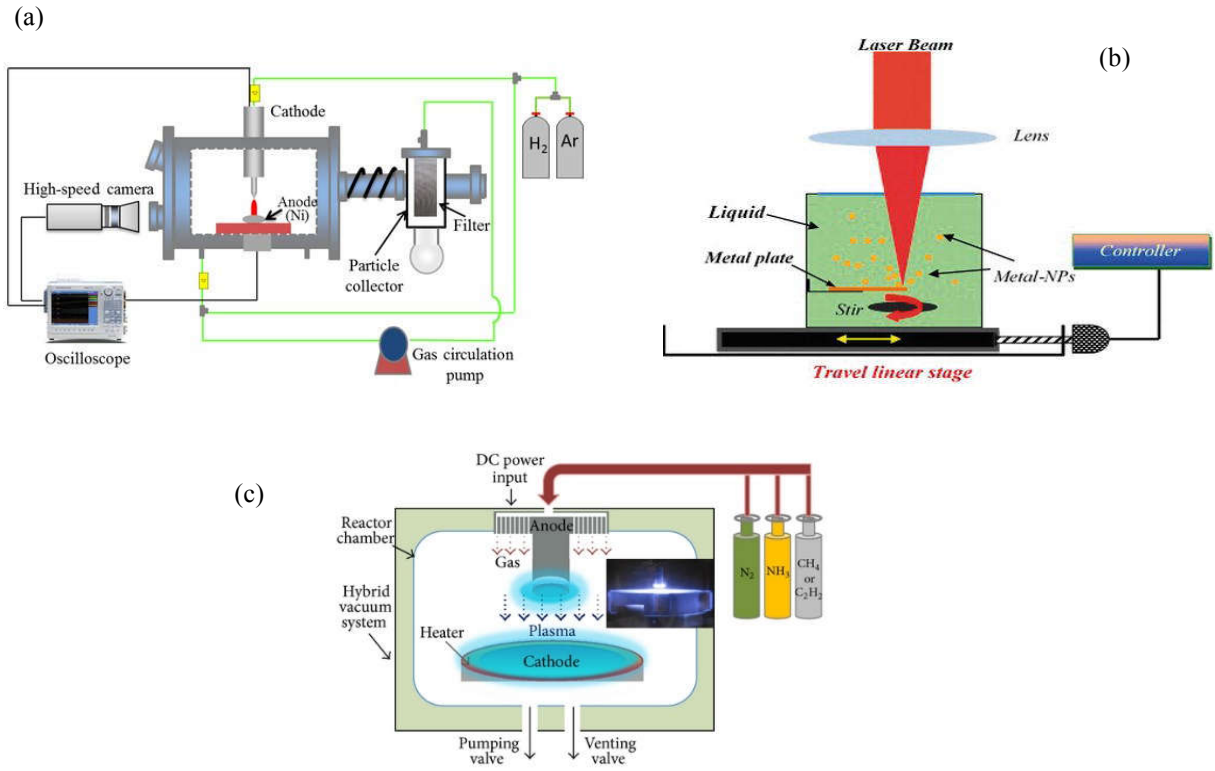

Figure 6: The most conventional CNTS synthesis techniques, (a) the arc discharge process, (b) laser ablation method, and (c) the chemical vapor deposition process. The attained figures are from open sources as [49] CC BY 4.0, [50] CC BY 3.0, and [51]CC BY 4.0, respectively.

CVD involves the decomposition of carbon species over a catalyst at relatively a high temperature to form CNTs. Among other synthesizing techniques (arc discharge and laser ablation), CVD is the most versatile technique for CNTs synthesizing. It is characterized by low production cost, high yield and the ability to produce different types of CNTs. Nowadays, CVD can be considered as the most trusted technique for mass production of CNTs[52].

6.2. CNTs for gas sensing:

The carbon nanotube is the most studied material for gas sensing applications. This is due to its high aspect ratio, chemical, thermal and mechanical stability, metallic and semi-conductive properties and functionalization capability that facilitate the adsorption of gas molecule on its surface. CNTs suffer from a change in theirconductivity as they interact with adsorbed gas molecules. Compared to conventional metal-oxide gas sensors, they have the advantage of detecting the presence of gases atroom temperature. However the pristine CNTs cannot differentiate between different gases, as they suffer from poor selectivity [53].Furthermore, the sensing mechanism of pristine CNTs is slightly complicated as they generally consist of a mixture of metallic and semiconducting tube, beside the different level of defects that resulted from purification processes.From this respect, modification/functionalization has been proposed to overcome poor sensitivity and selectivity [54]. Kinds of researches have been executed regarding the modification of CNTs with different materials. through this review, a summary of CNTs modified with inorganic an organic species will be exploited, especially forgas sensor devices with enhanced selectivity and sensitivity.

6.2.1. Carbon nanotubes/metal oxides for gas sensing

A variety of metal oxides semiconductors has been anchored to CNTs through different physical and chemical routes[55]. The main purpose of the decoration process is to promote the selectivity and sensitivity of CNTs for gas sensing applications. Both SWCNTs and MWCNTs have been studied extensively and some of the recent studies will be reviewed hereinafter. the SWCNTs are configured into gas sensors for a variety of poisonous gases like NH3, NO, and NO2. The SWNT-Fe2O3 composite film demonstrates a steady response and enhanced sensitivity for H2S and shows enhanced sensitivity to NO2 and at room temperature compared with pristine SWNT films. These flexible sensors with high deformability possibility can serve as wearable monitoring devices [56].

The combination of ZnO and SWCNTs is investigated for the detection of ethanol gas. The Synthesis process is the spray pyrolysis of the nanostructured materials on a copper substrate. The optimum device performance is displayed at a6% concentration of ZnO/SWCNT by weight. The chemisorption is the reason for the gas sensor response, in which the exchange of the charges between the metal oxide surface and the adsorbed gas species is attained[57]

The gas sensing for NO2 is also developed using the same SWCNTs coated ZnO prepared by wet chemical process, where the response and recovery times are 70 and 100 s consecutively. Best conditions for sensing performance are acquired for 1000 ppm NO2 at 150 ºC[58].

The electrochemical synthesis method is also conducted for the gas sensing evaluation using ZnO/SWCNTs hybrids. Room temperature performance for varied gases of CO, CO2, NO2, H2S, O2, and NH3 is executed. By finely adjusting the electrochemical factors, density, crystallinity, and subsequent particle size, a noticeable impact on the ambient conditions gas sensing attitude is monitored. In short, the functional ZnO/MWCNTs reveals almost 5% per ppm compared to 0.2 % for the non-functionalized MWCNTs for the H2S gas.

A combination of SnO2 and TiO_2 carbon nanotubes were prepared via a feasible sol-gel method for ethanol detection. The mixture demonstrates good quality as thermal stability and elevated sensitivity within a wide temperature span of room temperature to 250 ºC [59].The Exploitation physical properties and gaseous monitoring in MWCNTs filled with vanadium oxide was demonstrated. Methane gas detection response approached 16 seconds at room temperature, which was attributed to the density of states increase amongst Fermi energy level for the composite. Stress and highlights on the impact of ambient oxygen in CNTs-based sensors were addressed [60]. Cháfer etal explored the possibility of detecting both of NO2 and NH3. Using IrOx-MWCNTs nano composite. They demonstrated that IrOx-MWCNTs nano composite candetect NO2 and NH3 at different working temperatures with good reproducibility, higher sensitivity as low as 1bbp, stability, and lower noise levels in comparison to pristine MWCNTs [61].The MWCNTs doped ZnO is applied as a toluene gas sensor. Briefly, a variation of ZnO/MWCNTs is delivered which was synthesized by the reflux procedure. MWCNTs hinders agglomerations preparation for ZnO nanostructures. The ratio of 3:1 ZnO: MWCNTs reveals a 17% elevation of the sensor response at 150 ºC compared with the pristine ZnO that doesn't introduce any response at the mentioned temperature [62]. The room temperature operated ammonia gas sensor was developed by T. Guo et al. They utilized a CNTs/Fe3O4 composite to detect a low concentration of ammonia at room temperature. They subjected CNTs/Fe3O4 sensor for ammonia concentrations of 20, 40, 60, and 80 ppm. The resulted demonstrated that the sensor respond selectively for ammonia with a short recovery time. Beside these advantages, the sensor has good linearity, good repeatability and long term stability[63] as shown in fig. 7.

The MWCNTs decorated ZnO nanostructures are prepared for the hydrogen gas sensing. To enhance the performance, Pt nanoparticles deposited by sputtering are employed on the composite surface. 78 s recovery time is approached, as well as decent repeatability and stability for 0.05% amount of the targeted gas at surrounding room temperature. Moreover, the MWCNTs/ZnO/Pt reaches 4% sensitivity which is almost a duplication of the MWCNTs/ZnO itself [64].

The nanostructure of Al2O3/CeO2/MWCNTs was successfully prepared by the conventional chemical precipitation process under the impact of the ultrasonic wave for CO2 gas. The obtained results demonstrated that the thermal conductivity sensor has 9 and 13 s as a response and recovery durations subsequently which outcomes most of the commercially available CO2 sensors[65].The as prepared MWCNTs/ZnO by reflux method at 197 ºC in ethylene glycol is employed for methanol gas detection. A wide span of temperatures within 100-300 ºC was investigated for the sensing behavior where the best performance was gained at the latter

temperature. The Al2O3 doped with MWCNTs is also presented for methane detection [66].

6.2.2. Carbon nanotubes/metals for gas sensing:

Carbon nanotubes decorated with gold nanoparticles are examined for (NO2, CO, and C6H6) contaminants detection through a combination of experimental/theoretical routes. Gold nanoparticles depict a direct impact on NO2, and CO detection, where no significant effect was noticed in C6H6. This behavior discrepancy can be clarified by recognizing the connection between the change of resistance (macroscopic property) and Fermi level shift (microscopic feature) after gas adsorption [67]. Additionally, to these materials carbon nanotubes were decorated with rhodium nanoparticles and served as a sensor for the detection of f NO2, C2H4, CO, C6H6.Oxygen existence is crucial for the enhancement of gas response because the oxygenated vacancies behave as active adsorption locations for gases and as anchoring sites for rhodium nanoparticles. Additionally easier charge transmission between rhodium nanoparticles and carbon nanotubes can be achieved [68]. Sharafeldin etal., decorated the MWCNTs with Cu, Pt, Ti, Ru and Ag to investigate its gas sensing behavior. They found that the MWCNTs/Cu nanocomposites exhibited the highest sensitivity of 1.75% when exposed to 10ppm H2S. Also they demonstrated that the MWCNTs/Pt exhibited a greater response of 1.96 % during exposure to 10 ppm NO2[69]. The zigzag MWCNTs coated with Pd and Pt nano particles were exposed to different concentrations of CO and NO at room temperature. This novel structure was studied employing first-principles calculations. The deep analysis showed that the Pt decorated SWCNTs are more sensitive to CO while, the SWCNTs decorated with Pd is highly sensitive to NO [70]. The Au nanoparticles with controlled size and ratio was developed over MWCNTs to establish an NO2 gas sensor to detect minor concentration of down to sub ppm level. This study concluded that the deposited amount of Au nano particles control the sensing capabilities. The low Au loading, the heights response toward H2S [71]. Khan et al., was utilized the simple and cost-effective spray method for depositing Ag/CNTs and ZnO/Ag/CNTs over a cellulose paper. They demonstrated that the Ag/CNTs sensor respondedselectivelyfor acetone fasterCNTs/ZnO/Ag sensor [72]. The CNTs functionalized with Ni nanoparticles was adapted to detect SO2, H2S, and SO2F2. Y. Gui et al., reported that the low detection limit (LOD) of the Ni/CNTs sensor was 1 ppm against SF6. They demonstrated that the sensitivity toward different gases in the following order H2S > SOF2 > SO2 > SO2F2. [73]

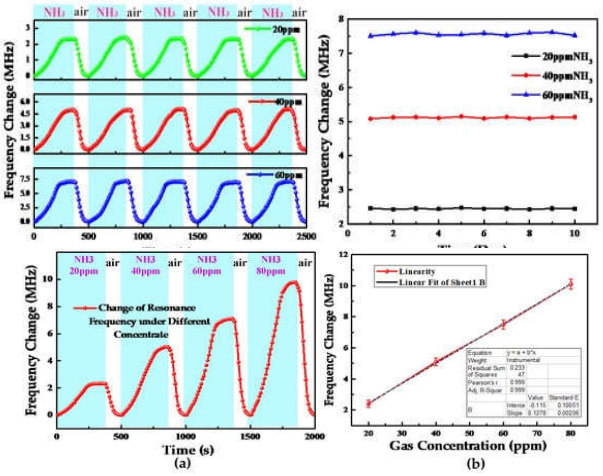

Figure 7: CNTs/Fe_3O_4 gas sensor a) repeatability, b) Stability, c) response at different concentration, and d) linearity, [63]CC BY 4.0

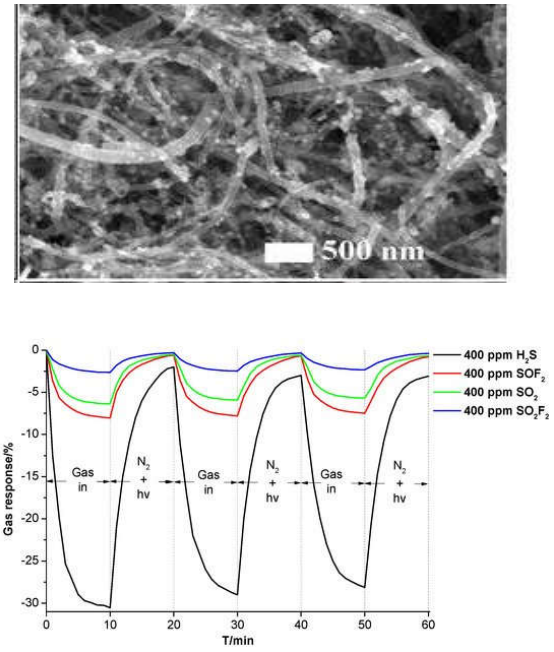

Figure 8:a) HRTEM of CNTs decorated with Ni nano particles and b) the response of CNTs/Ni sensor toward different gases. This figure is obtained from [74] CC BY 4.0

6.2.3. Carbon nanotubes/ organic materials for gas sensing

CNTs/Poly-ethylene glycol composite was used as gas sensors to detect volatile organic compounds (VOCs). Reachable high response (≈110 s) and recovery (≈152 s) rates were acquired in diverse concentrations of acetone, ethanol, isopropanol and isoprene at ambient temperature. These rates

nominate the sensor as a portable electronic-nose device [75]. Gas sensing can also be promoted based on poly (3,4-ethylenedioxythiophene) polystyrene sulfonate – multiwall carbon nanotubes (PEDOT: PSS-MWCNTs) as the sensitive material. This work targets the development of a low-cost communicating sensor that attain the possibility of sensing platform integrationdevoted to low power applications as well as the Internet of Things (IoT) [76]. Gas sensing of NO2 is utilized based on single walled nanotubes mounted on polytetrafluoroethylene (PTFE) filter flexible substrates. A demonstrated stability of sensitivity was depicted during repeated bending of the substrates within 0.75–2 ppm concentrations. However, for 3–5 ppm concentration, a remarkable rise in the sensitivity was achieved. This can be linked to the porous nature of the substrates. Compared with the sensors fabricated over a silicon substrate, duplication of sensitivity can be obtained. Moreover, a decrease of the sensitivity at 10 % and 30 % humidity because of the electron donor nature of water molecules. These results are beneficial for flexible electronics and for monitoring air quality [77].

The detection of ammonia gas is conducted relied on CNTs and polyaniline films. Three different doping routes were performed (i.e. sulfuric acid, camphorsulfonic acid and m-cresol). The optimum sensor response was acquired for the camphorsulfonic acid is comparable with others. This can be regarded to the conservation of polyaniline primary volume, and the well-distributed polarons initiated by this doping agent. This device operates with an optimized sensitivity for ammonia gas detection with a limit of detection of 4 ppm [78]. Chiou et al., demonstrated an acetone gas sensor of polyethylene glycol (PEG)/MWCNTs composite film. The results for sensing performance revealed a higher sensitivity for PEG/MWCNTs in the presence of moderate temperature than the absence of the thermal treatment which is beneficial for environmental applications [79]. W. Zhangetal., explored the efficacy of PANI/CNT composite for the monitoring of NO2 and NH3. They improved the sensing performance of PANI/CNT composite via establishing a core-shell structure ofp-type MWCNTs and n-type PANI. The low detection limit (LOD) of their structure was attained as 19.6 and 6.5 ppb, for NO2 and NH3 respectively as illustrated in fig. 9[80]

Recently, a variety of 3,4 ethylenedioxythiophene)/poly (4-styrenesulfonate) concentrations were examined for the application of 3D 3,4 ethylenedioxythiophene)/poly (4-styrenesulfonate) (PEDOT: PSS) multi-walled composite carbon nanotubes in gas sensing, where using ethanol and CO. Enhanced response and recovery times as well as increased conductivity were observed for ethanol and CO. From one side the gas molecules adsorption sites reduced with PEDOT: PSS coating layer, but from the other the charge-carrier transport increases within the multi-walled carbon nanotubes and gas molecules, which is translated finally to a better 3D networking and potential applicability for gas sensing [81].

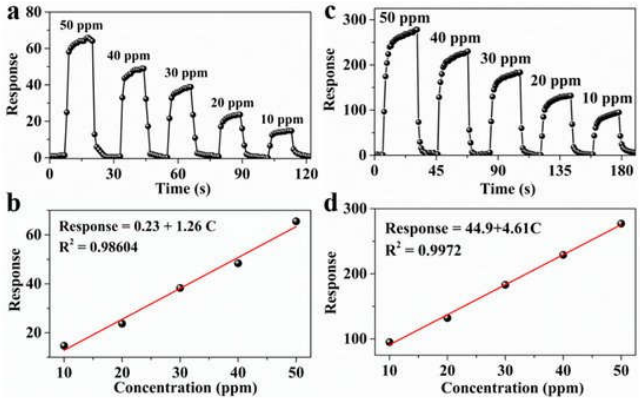

Figure9:(a) The response of p-PANI/CNT toward NO_2 gas , (b) the fitting curve of sensor p-PANI/CNT against NO_2 concentration, (c) responses of p-PANI/CNT toward NH_3 gas, (d) the fitting curve of sensor p-PANI/CNT against NH_3 concentration, The figure is taken from [80] CC BY 4.0

6.4. Graphene

Graphene is a unique two dimensional material with a honeycomb crystal lattice,introduced initially by Novoselov et al[82], existing in graphite, coal, and created in other meansIt owes a distinctivespecification such as flexibility, increased surface area, high mobilityand mechanicalstrength,environmental steadiness, versatility, and optical transparency. Even in a vacuum, it maintains high mobility which outcomes Si and Cu. Thanks to the conferred aforementioned properties, graphene is avery promising material for a range of industrial applications, whichare engaged innanotechnological areas as biosensing[83, 84][85][86], water treatment [87], photocatalysis and solar cells [88],[89], semiconductors [90][59], energy storagedevices [60], as well as biomedical sector[61][62].Moreover, graphene is the standard building block for nanotubes, and 3D graphite. The resulted graphene from graphene oxide reduction differs in structure from pristine graphene. While the former has wrinkled structure owing to lattice defects existence, the latter displays a rippled structure [63]. Graphene can be created by mechanical or chemical

exfoliation. Other processes rely on chemical vapor deposition and other methodologies[64].

6.4.1. Synthesis of graphene

Graphene deserves great attention as it has a large theoretical specific surface area (2630 m2g-1), high intrinsic mobility (200,000 cm2 v-1s-1), [65][66] high Young's modulus (*1.0 TPa) [67]and thermal conductivity (5000 Wm-1K-1), [68][69][70][71]optical transmittance (*97.7 %) and good electrical conductivity, on other hand, preparation of the large specific surface area, high quality is not applicable in a low cost-effective way.

Monoatomic or single-layer graphene is called one graphitic layer, also by the same way bilayer graphene contains two graphitic layers, and tri-layer graphene contains three graphitic layers. Few layer graphene referred to more than 5 layers up to 10 layer graphene, and multilayer graphene, nanocrystalline, or thin graphite thick graphene is pointed to *20–30 layer graphene.[72]. for over 40 years graphene has been developed [73–79] and measurements of properties in exfoliated layers [80], of graphene grown on (Si) [81], large-area graphene grown on copper (Cu) substrates [82], as well as different research including the involvement of chemically modified graphene (CMG) to produce novel, and modern materials. [77–86]

Graphene can be synthesized by many methods through mechanical cleaving (exfoliation), chemical exfoliation, chemical synthesis (reduced graphene oxide), and thermal chemical vapor deposition (CVD), which are the most generally used nowadays. A flowchart of graphene preparation techniques is obtained in Fig.10.

6.3.1.1. Mechanical exfoliation

Mechanical exfoliation is the most extraordinary and prominent technique for preparing single-layer graphene. Graphene synthesis can be synthesized through mechanical exfoliation as the first method used. mechanical exfoliation is a top-down technique, by which linear or transverse stress is generated on the surface of the flaked structure materials.

For mechanical exfoliation, the external force required to separate one mono-atomic layer from graphite is 300 nN/cm2. [87]

Actually various thicknesses of graphene flakes could be created by mechanical exfoliation or exfoliating sheets from graphitic materials like highly ordered pyrolytic graphite (HOPG), natural graphite, or single-crystal graphite [88][89][90][78][91][92]

This exfoliation can be performed by different types of agents such as scotch tape [80], ultra sonication, [93]electric field [94],and even by transfer printing technique, [95][96],etc.

In this micromechanical exfoliation method, graphene is separated from a graphite crystal using adhesive tape.

Actually it is not easy to obtain larger amounts of graphene by this exfoliation method, not even taking into account the lack of sustainable flakes. The difficulty of this method is low, nevertheless, the graphene flakes require to be found on the substrate surface, which is labor exhaustive. The quality of the prepared graphene is very high with almost no defects. Still, the mechanical exfoliation method needs to be enhanced further for large-scale, defect-free, high-purity graphene for mass production in the field of nanotechnology.

6.3.1.2. Chemical exfoliation

The chemical method is one of the best appropriate methods for the synthesis of graphene[130]. In chemical method producing colloidal suspension which modifies graphene from graphite and graphite intercalation compound. Different types of paper-like material [85][97][98][99][100][101]polymer composites,[83] energy storage materials [102]and transparent conductive electrodes [103]have already used chemical methods for production of graphene. Chemical exfoliation is a two-step process. At first, reduces the interlayer van der Waals forcesto increase the interlayer spacing. Thus it forms graphene intercalated compounds (GICs) [104]. Then it exfoliates graphene with a single to few layers by rapid heating or sonication as schematically represented in fig. 11.

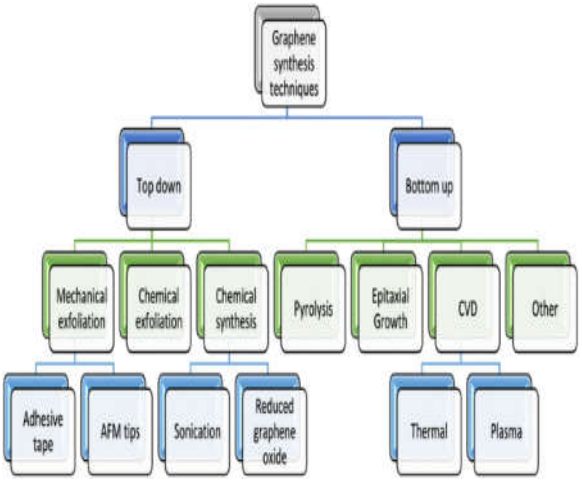

Figure 10: an overview of the most common routes for the graphene preparation methods, source[119] CC BY 4.0

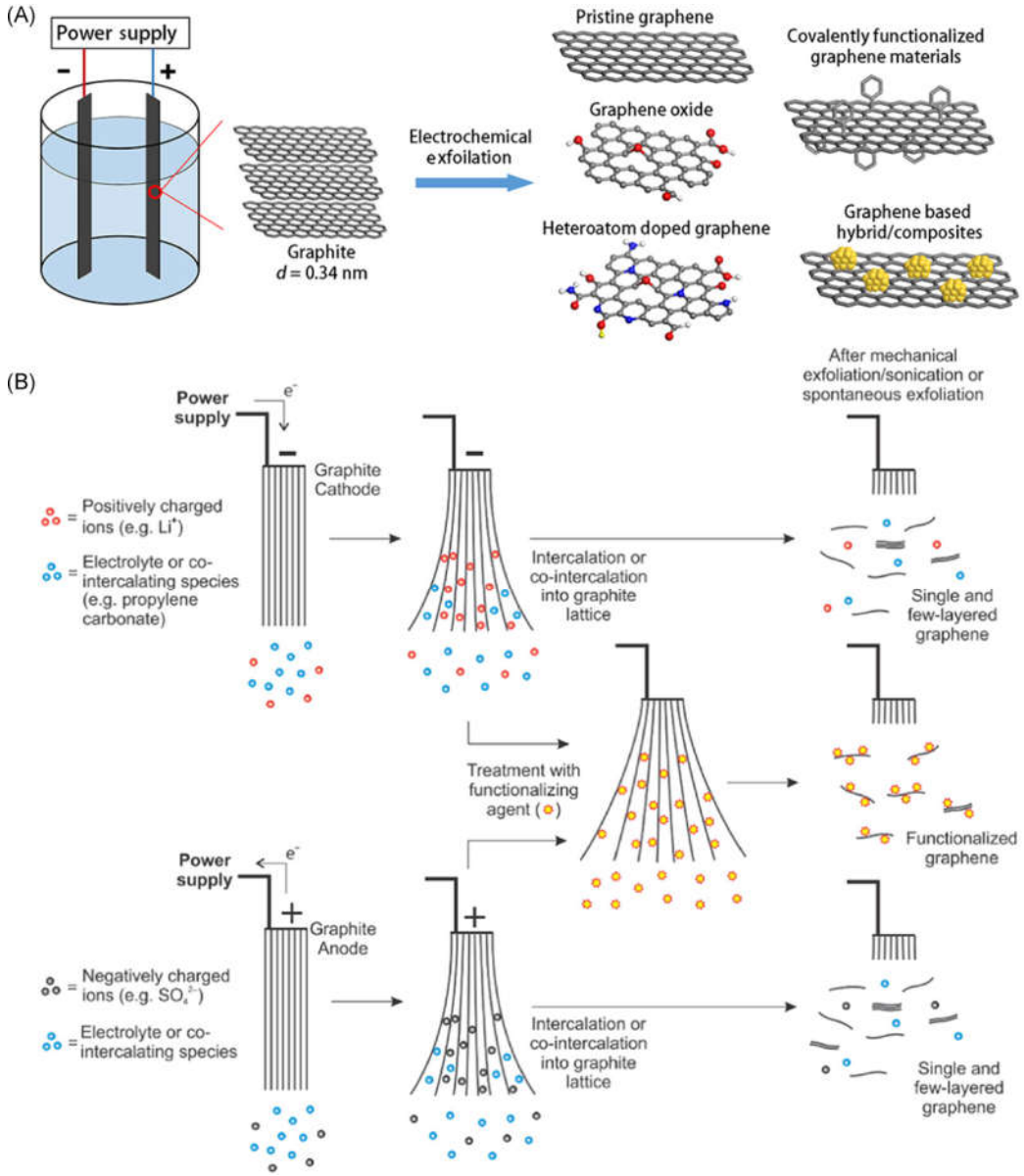

Figure 11: The representation of the chemical exfoliation of the graphene, source: [139] CC BY 4.0

6.3.1.3. Pyrolysis of graphene

One of the chemical methods forthe synthesis of graphene in the bottom-up process was the solvothermal method. The molar ratio of sodium and ethanol was 1:1 in a closed vessel in this thermal reaction

Graphene sheets could be in a smooth way pyrolyzed of sodium ethoxide using sonication. These graphene sheets were produced with dimensions of up to10 µm. The crystalline structure, different layers, graphitic nature, band structure were established by TEM and Raman spectroscopy [105].

The benefits package of thepyrolization were low-cost and easily fabricated of high-purity, functionalized graphene at low temperature. Yet, the modality of graphene sheets was still not suitable because it comprised a large number of imperfections.

6.3.1.4. Epitaxial growth of graphene

Epitaxial thermal growth on a single crystalline silicon carbide (SiC) surface is one of the most praised methods of graphene synthesis. The schematic representation of the process is depicted in fig. 12.Silicon carbide (SiC)was first used in electrical measurements of patterned epitaxial graphene [80]. Epitaxial graphene growth on SiC has been visualized as a very promising method for large-scale production and commercialization of graphene for applications into electronics. Graphene on SiC produces high-frequency electronics [106], light-emitting devices [106], and radiation hard devices [106]. Top gated transistors have been fabricated from graphene on SiC on a wafer-scale [107]. High-frequency transistors have also been revealed with 100 GHz cut-off frequency208 [143], higher than state-of-the art Si transistors of the same gate length. Graphene on SiC has been established as a novel resistance standard based on the quantum Hall effect (QHE) [109][82]. Though this process is very expensive.

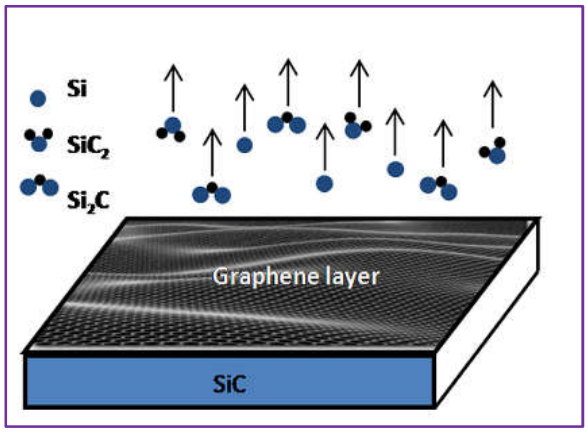

Figure 12: The epitaxial growth of graphene on SiC substrate, source: [145], CC BY 4.0

6.3.1.5. Chemical vapor deposition (CVD)

The deposition of high-quality graphene from the CVD process is usually done onto various transition-metal substrates like Ni [110] Pd [98], Ru [111], Ir [112], and Cu [113]. CVD growth of graphene has been mainly practiced on copper [113][114] and nickel [113][110] substrates. Nickel was the first substrate on which CVD growth of large-area graphene was attempted. These efforts had begun right from 2008. [115].

Different hydrocarbons such as methane, ethylene, acetylene, and benzene were decomposed on various transition metal substrates such as Ni, Cu, Co, Au, and Ru [113]. Single crystals using an ethylene precursor were found to yield graphene structurally coherent even over the Ir step edges [112].

Depending on the material quality, precursors, width, and the structure required; there are many various types of CVD processes: thermal, plasma enhanced (PECVD), cold wall, hot wall, reactive, and so on.

In CVD process reactors like hot-wall reactor, there temperature is relatively constant everywhere and these walls never get heated in the cold wall systems. Graphene is formed on Cu thin film mostly by cold wall system.

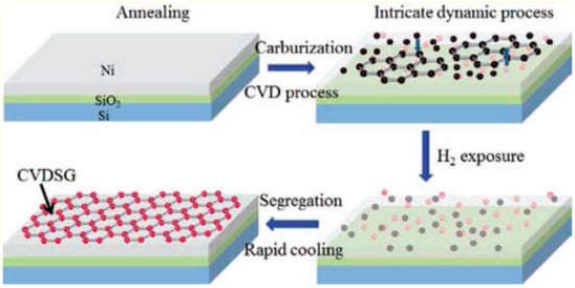

Figure 13: The elucidation of the CVD growth of graphene, source:[152] CC BY 4.0

6.4.2. Graphene/ polymers for gas sensing:

The monitoring and detection of nitrogen dioxide were utilized by employing the rGO/polymer nanofibers. Room temperature applicability and high sensitivity of 1.03 ppmwere feasiblyacquired. Moreover, 150 ppm detection limit was achieved, synthesis methodology using electro-spun offers a feasible, eco-friendly and robust route for preparation[116]. The acquisition of ammonia sensors was proposed relied on the rGO/ conductive polymers combination through the Langmuir-Schaefer (LS) procedure. The implication of exploiting pyrrole (Py) as the reducing agents demonstrated thehighest sensitivity for pyrrole-rGO-polyaniline amongst the four synthesized samples. A detection limit of 0.2 ppm was maintained[117]. Graphene/ethyl cellulose nanocomposite was introduced for the highly sensitive wearable gas sensor with reduced strain response, this sensor displays0.3% comparative resistance variation at thesmallest bending radius of 3.18 mm after 400 bending cycles. For a bending radius of 5 mm, 0.2 % resistance change was grasped. Detection limits rangefrom 37 to 167 ppm was also monitored. The detection of ethanol, acetone, IPA, and hexane were reported[118].

6.4.3. Graphene / metal oxide nanocomposite for gas sensing:

Metal oxides like ZnO, MnO2,WO3, MoO3and CuO are broadly investigated for sensing applications [119][120][121][55][159][45], from one side they have raised specific area for the surface and decent flexibility [122]from the other side they lack the satisfactory electrical conductivity. The gathering of graphene and its derivatives with the metal oxides can inherently enhance the electrical conductivity and the sensing performancesubsequently. The introduction of graphene/metal oxide is reported[123].The incorporation of metal oxides onto graphene results in emerged physical and chemical specifications. Moreover it possessesa vital role in the suppression of graphene sheets of aggregation [54].The nanocomposite of NiO/ rGO was introduced for the detection and sensing of methanethrough the hydrothermal method. The probable mechanismof sensing was credited to the Fermi energy band among NiO nanoparticles and rGO sheets. High response times of approximately 6-18s were noticed for 100-500 ppm concentrations[124].The detection of formaldehyde is pursued relied on another metal oxide (i.e. ZnO)-rGO nanocomposite by Weiwei Guo et al., where the ZnO was doped by Fe and the nanocomposite was prepared via a one-pot hydrothermal route. The incorporation of Fe onto the ZnO-rGO nanocomposite results in a reduction in the ZnO hexagonal prism accompanied by an elevation of the surface area for the aforementioned ZnO. A ratio of 5 % doping of Fe sensing performance maintains a response-recovery times of 34, 37 seconds for 12-5 ppm concentrationof formaldehyde[125]. The same ZnO modified with rGO is employed for the ultrasensitive monitoring of NO2 gas. Seven-fold enhancement of the response compared to pristine ZnO at 100 ºC is approached. Detection limit as low as 5ppm is attained. The elevated performance of the sensor is regarded to the existence of p-n heterojunctions amongst the rGO and ZnO[126].The hydrothermal method for preparation was introduced to acquire the rGO-TiO2 nanocomposite with the objective of ammonia sensing. The mentioned nanocomposite demonstrated enhanced selectivity and sensitivity for ammonia concentration down to 5 ppm at room temperature[127]. While the same nanocomposite (rGO-TiO2 nanocomposite) can detect the CO gas for reduced 100 ppm concentrations[128]. Vardan Galstyan et al., fabricated the rGO/ZnO nanocomposite for detecting NO2, H2 and CH4 gases. the incorporation of rGO enhances the conductivity of the resultant composite, thereby promoting the response to NO2, and H2 gases as shown in fig. 14. compared to pure ZnO, the rGO/ZnO nanocomposite respondsselectively to to NO2, gas at relatively low working temperature[167]

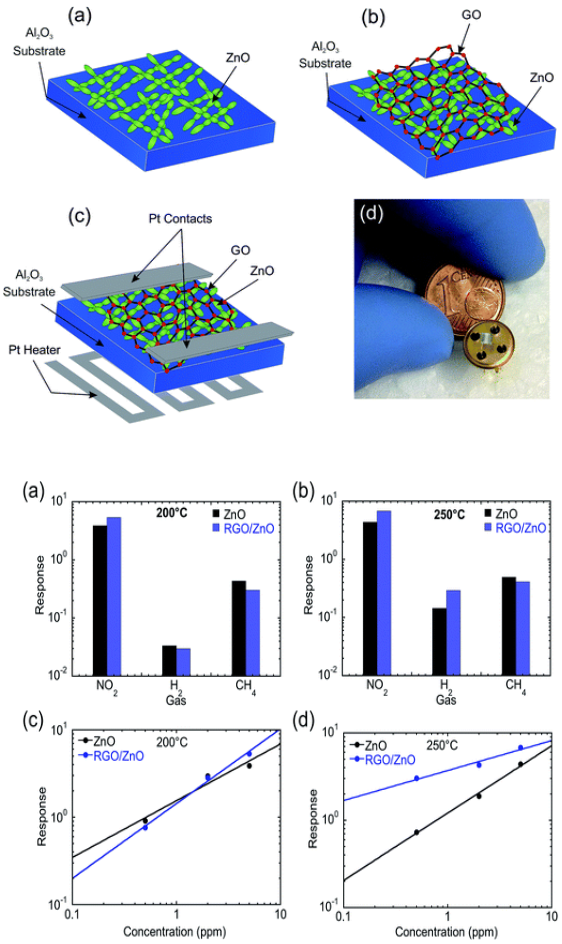

Figure 14: The schematic representation of gas sensor device fabrication.Ref. [167] open access

The detection of another gas (NO2 in this case), is demonstrated using GO-WO3 nanocomposite films. The synthesis was performed by a combination of the polyol process and metal-organic decomposition. Imminent sensitivity within 0.5–5 ppm with nice reproducibility was affirmed. Long-term stability for more than a month is reported at room temperature[129].The decoration of copper oxide with reduced graphene oxide was exploited for carbon monoxide sensing through the layer-by-layerself-assembly methodology. A spacious range of 0.25 ppm to 1000 ppm CO concentration was investigated. Excellent performance for reproducibility, sensitivity, and stability was demonstrated, which originated from the formulated heterojunction at the CuO-rGO interface[130]. The hybrid nanocomposite of SnO2 and GO revealedrecognized sensitivity to detect NH3with 10 to 50 ppm at ambient temperature[131]. The carbon

monoxide sensing is also attained using the rGO-Fe3O4 nanocomposite, the response-recovery timings of 32-35 s are acquired respectively for 5 ppm concentration[132].Another type of CO gas sensing for NiO/grapheneusing the hydrothermal reflux method was introduced[133]. The composites of nitrogen doping onto graphene decorated with MnO2 was prepared by a means of microwave-assisted procedure. The microwave irradiation facilitated the development of nanoflowers because of the inherent interactions amongst the alternation current and the electric dipoles[134]. The tin oxide modified reduced graphene oxide (SnO2-rGO) nanocomposite was prepared through one-step hydrothermal procedure for the monitoring of H2S and SOF2. The optimal conditions outcomes the pristine rGO sensors. Acquired elevated responses of (34.31% and −3.13%) higher than tan those (5.97% and −1.45%) of pristine rGO sensor at 125 °C, for 100 ppm H2S and 10 ppm SOF2, concentrationscorrespondingly[135]. A Tri-structure system of rGO-SnO2-Au was effectively synthesized to detect the formaldehyde. Remarkable higher sensor response and selectivity could be attained. This enhanced response might be attributed to the fact of ohmic contact synergistic effect between SnO2 and rGO, as well as the increased surface area, and Au nanoparticles catalytic effect. [136].

6.4.4. Graphene / metals nanocomposite for gas sensing

Graphene-noble metal gas sensing was also introduced earlier [137][138]Recently, GO- metals are of great interest thanks to their enhanced catalytic, electronic and optical specifications[139].The hydrogen gas sensing is performed using the Pt-Pd/rGO. Stability and repeatability for the response are confirmed. This might be attributed to the carrier donation and the expansion in the crystal lattice during hydrogenation and dehydrogenation. The response can be enhanced by increasing the hydrogen concentration or reducing the operating temperature. The flow rate alteration doesn't reflect any noticeable changes in the sensor outcome. Upon employing nitrogen instead of air as a gas carrier, higher response/recovery durations were acquired. This might be regarded to the contribution of oxygen in the reaction[140].NO2 sensing is also recalled by introducing the impact of electron beam irradiation for Pd-fortified rGO composites. Altered doses of irradiation were executed from 0 to 500 kGy, where the latter dose demonstrates the optimum response. The response time of 345 s was acquired for a 10 ppm concentration of NO2, while the recovery time hits 816 s for the same concentration and dose. Oxygen functional groups abundance along with high energy defects initiates this elevated gas response[141].

Pd/rGO hybrid, is used to sense the hydrogen gas. The hybrid was synthesized via microwave irradiated assisted route. Sensing performance was investigated through a spacious temperature range- from room temperature to 120 ºC. The highest response of 14.5 % was achieved for 1 % H2 sensing at 100 ºC. this remark might be attributed to the raised amount of hydrogen molecules that interact with the sensing layer[142]. Ammonia gas is also detected using the rGO samples decorated by Ag, Au, and Pt nanocomposites synthesized by a single-step chemical reduction process. Amongst these three nanoparticles, silver demonstrated the highest recovery, response, and sensitivity [143]. In recent work, the detection of NO2 employing Au ornamented porous structure graphene was achieved at the normal room temperature. The sensor maintains a response to diminished gas concentrations as low as 50×10-9 within30 s. Au decoration elevated the sensitivity to ~1.5 times higher than pristine graphene[144]. The use of the ternary composite of Ag-MoSe2/reduced graphene oxide (rGO) for H2S gas sensing, where the composite was fabricated by following the hydrothermal process. The investigation of varied concentrations 0.1 ppm–30 ppm at room temperature was performed. The loading of Ag onto the compound has an apparent impact, which was regarded to the modulation of the potential barrier during electrons transfer, as well as the synergistic consequences of the ternary composite structure[145].

6.4.5. Graphene / CNTs/ Metal oxide nanocomposites

The nanocomposite of the graphene derivatives and carbon nanotubes mixed with metal oxide for usage in gas sensing applications is also investigated[185]. In addition to these composites carbon nanotubes/ reduced graphene oxide NO2 gas sensors are introduced. These sensors are supported by flexible polyamide substrate and operated at room temperature. These sensors are remarked with high sensitivity and high bending ability, where the former is due to CNTs array existence and the latter is regarded to the excellent flexibility of graphene films [146].

M. Morsy et al., recently reported that the mentioned nanocomposite incorporated with ZnO acquired by the conventional precipitation procedure benefits in the detection of ammonia gas at room temperature. Increased response and recovery times were noticed for the detected gas under investigation[147]. Inherently, the

graphene/SnO2prepared using the sol-gel method has a better gas sensing response than the graphene/MWCNTs/SnO2 at room temperature for NO2as reported by Vibha Srivastavaet al.,The composite is signified by a reduced time of response < 1min, and almost 5 min for the recovery period. The raised response can be regarded to the full exposure of the surface towards the surrounding environment [188].

6.4.6. Three-dimensional graphene and graphene foam.

The three-dimensional (3D) graphene foams [148]are introduced as well for different application areas such as Li batteries[149], photocatalysis [150] water treatment [151] and else[152][153].The freeze-drying (FD) procedure is used to produce 3D graphene nanoplatelets (GNP) foam. This method introduces better mechanical specifications which outcomes the CVD graphene foam. However, it possesses 0.5 times thermal diffusivity because ofdue to the pore architecture and defects in GNP particles[154].The detection of glucose is also introduced using graphene foam based as an electrochemical sensing approach. Macroporous 3D graphenefoam synthesized by chemical vapor deposition (CVD) helped as the electrode scaffold. The reduced detection limit of 1.5 μM is obtained within a spacious linear range (5-65 μM)[155]. In another report, the macro graphene foam- like is employed for the detection of gases. Herein the combination between the high sensitivity and appropriate reliability. Few ppm levels were attained for the NH3 and NO2 gases detection in the air at the ambient situation. Furthermore, the proposed combination attains a desirable mechanical strength and flexibility as well as reduced power consumption regarded to Joule-heating which opposes chemisorbed molecules from the surface of the foam [156]. The 3D reduced graphene oxide (3DRGO) decorated with ZnO nanoparticles was investigated to detect the CO gas. The studied performed by N. HaiHa et al., confirmed the 3DRGO/ZnO composite based gas sensor has a fast response and recovery, good linearity, stability, and enhanced selectivity [198]. The detection of VOCs was execute by 3Dgraphene loaded with Co3O4. The GF/Co3O4 nanocomposite exibeted a high response and fast response time toward Xylene at low concentrations [199].

Conclusion:

In this review article, we have surveyed the gas sensing implications for versatile carbon-based nanomaterials. Initially, we have started with an introduction which presents carbon nanohorns and nanoonions, subsequently heading to nanodiamonds, and carbon quantum dots. At the end of this section, we finish with the carbon nanotubes (CNTs). In the second section, the research progress for CNTs versatile combinations is inherently explored. Herein, the gas sensing performance and evolution for CNTs/metal oxides, CNTs/metals, and CNTs/organic materials highlighting the enhanced detection limits and response/recovery timings are included. Thirdly, we shed lights on the graphene synthesis protocols such as mechanical and chemical exfoliation as well as chemical vapor deposition (CVD). Finally, we investigate graphene combinations for gas sensing. This comprises graphene/metal oxide, graphene/ noble metals, and graphene/CNTs/metal oxide. The propagation of the discussion from 0D. 1D to 2D and ending with 3D foam graphene is involved. This gas sensing review delivers the keypoints for the selection of the appropriate carbon-based materials and the accompanying implications needed for the next era of technological and industrial applications.

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الملخص العربى

المواد الكربونيه النانومتريه ومتراكباتها وتطبيقها كحساسات للغاز : مقاله مرجعيه

اثارت النتائج المترتبه على دمج المواد الكربونيه فى المدى النانومترى لتطبيقات حساسات الغاز الاهتمام فى السنوات القريبه المنقضيه. طبقا للخواص الفيزيائيه والكيميائيه المرغوبه للمركبات الكربونيه النانومتريه ومتراكباتها المختلفه فانها تتناسب مع تطبيقات حساسات الغاز حيث انها تستوفى شرط مساحة السطح الكبيره وكذلك فجوة الطاقه المنخفضه. فى هذه المقاله المرجعيه نبدأ باستعراض اهمية وطرق تحضير المواد الكربونيه للتركيبات المتنوعه مثل (carbon nanoonions), (carbon naohorns), (nanodiamonds), and (carbon quantum dots)الجرافين ومشتقاته المتنوعه يتم دمجها عامة مع انابيب الكربون النانومتريه. فى الجزء التالى لما سبق نتطرق الى طرق تحضير الجرافين وخصائصه ،ومتراكبات الجرافين-الاكاسيد المعدنيه ،و الجرافين - العناصر النبيله ،و الجرافين-المواد البوليمريه المطبقه لحساسات الغاز. هذه المقاله المرجعيه تلخص النقاط الاساسيه والمؤثره لتطبيقات حساسات الغاز المعتمده على المواد الكربونيه فى المدى النانومترى. اضافة لما سبق فانه تم مناقشة واستعراض حساسات الغاز لانواع مختلفه من الغازات.