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Review

Carbon and graphene based nanocomposites for gas sensors—Current state and advances

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Abstract: After the discovery of carbon nanoforms, carbon nanotube (one dimensional) and tube like nanostructure and graphene (two dimensional) nanosheets have gained immense research curiosity. Further nanotechnological developments have moved towards the formation of carbon nanotube nanocomposites and graphene nanocomposites. For the purpose, various matrices including thermoplastic polymers and conjugated polymers have been used. Methodology is the systematic gathering of the literature and development of a novel review outline, theme, and discussions regarding the discussed topics. Hence, varying conjugated polymers such as polyaniline, polythiophene, poly(3,4-ethylenedioxythiophene), and nonconjugated nylon, poly(ethylene glycol), etc. have been processed using techniques like in situ, solution, electropolymerization, spin coating, etc. In sensors, the nanocomposites need to develop fine nanoparticle dispersion, network formation, and interfacial interactions ultimately supporting the electron or charge transfer in these nanomaterials desirable for the recognition of the gaseous species. Moreover, interactions of the nanocomposite with the analyte molecules define the sensing capabilities of the nanomaterials. Consequently, nanocarbon nanocomposite based gas sensors have been analyzed for conductivity, change in resistance, sensitivity, selectivity, response time, detection limit, and other desirable properties. For future designs, it is recommended to develop high-tech combinations of conjugated polymers like polythiophene derivatives using functional forms of graphene and carbon nanotube. In addition, use of advanced manufacturing techniques like 3D/4D printing and spin coating must be applied to form efficient sensors. In conclusions, this manuscript presents not only comprehensive but also comparative analysis on different gas analysis parameters such as detection limit, concentration, response time, etc. for various nanocomposite sensors. Lastly, the encounters in preparing and applying graphene/carbon nanotube sensors, associated utilizations, and possible future prospects have been discussed.

Keywords: carbon nanotube; graphene; polymer; nanocomposite; conductivity; gas sensing

1. Introduction

In the field of sensors, efficient carbon nanomaterials have gained interest [1]. Accordingly, the advanced sensing features have been observed for the nanocomposites [2]. Different types of matrices including conducting and non-conducting matrices have been applied in gas sensors [3–5]. Non-conductive matrices include polyamides and olefinic polymers to form the sensing nanocomposites [6]. Conjugated polymers for gas sensors include polyaniline, polypyrrole, polythiophene, and related polymers [7]. Forming nanocomposites of carbon nanoparticles with these matrices have been found to upsurge the electron conduction through formation of percolating and interconnected network nanostructures [8,9]. Carbon nanoparticles like graphene, carbon nanotube, carbon nanofibers, etc., in nanocomposite form have revealed fine sensing features towards gases, ions, and chemical species [10]. These nanomaterials have been designed through facile techniques like electropolymerization, in situ, solution, coating, etc. [11,12]. Such nanocarbon nanocomposite designs have been reported for better molecular recognition, detection, and rapid responses for the desired analytes in addition to fine electrical, dielectric, electrochemical, and other structural properties [13,14]. For enhanced sensing properties, carbon nanostructures have known to develop the charge transfer complex due to interactions and so revealed better microstructure, charge transport, and specific interactions for analytes causing high responsiveness and detection limit [15].

This state-of-the-art article covers gas sensing designs and features of the systems based on carbon nanotube nanocomposites and graphene nanocomposites. The multifunctional nanocarbon nanocomposites in gas sensing revealed high sensitivity, selectivity, and response time values. This overview portrays the progress in the field of two important nanocarbon nanostructures for gas sensing applications. Here, the nanocarbon nanofillers have been used in combination with important polymer matrices and sensing behavior has been analyzed for various gaseous species. Hence, this comprehensive review reports on the fundamentals to advanced potential of carbon nanocomposite sensors. Methodology of this review involves systematic gathering of scientific information collected from the reported literature on graphene and carbon nanotube nanocomposites. Consequently, the applications of these nanocomposite systems have been observed for the gas sensing application. During this review development, novelty is particularly considered in terms of the literature discussed, outlined topics, and variation of nanocomposite types, and polymers used for the formation of nanocomposite sensors. Hence, purpose is to report a radical and up-to-date article on nanocomposite gas sensors portraying indispensable features from fabrication—to—advanced potential. Need of developing this review has been analyzed due to the lack of comprehensive recent review articles in the field of carbon and graphene filler nanocomposite for gas sensing purposes. Although research reports can be seen in literature, however an all-inclusive article throwing light on the past, current, and predicted future developments need to be developed to benefit the interested field experts. Future progress in this field is not possible for the researchers before getting prior knowledge of the gathered literature on these nanomaterials. Hence, current state, advancements, future, and challenges in the field of nanocarbon based gas sensors have been comprehensively deliberated. This article is definitely beneficial for the field researchers and scientists striving to investigate better sensing designs in the field of nanocarbon sensors.

2. Gas sensing potential of carbon nanotube nanocomposites

Carbon nanotube was initially discovered in 1991 [16]. Since that time, carbon nanotube has been applied in significant industrial arenas [17]. Carbon nanotube is a hollow one dimensional nanotube like nanocarbon nanostructure consisting of sp^2 hybridization [18,19]. Carbon nanotube can be categorized as single walled, double walled, as well as multi walled nanostructures [20,21]. A single walled carbon nanotube has 1 nm diameter and around 100 nm length with chiral features [22]. Cabon nanotube has been analyzed for outstanding optical, electrical, mechanical, and physical properties [23]. Synthesis of carbon nanotube has been performed using variety of techniques like chemical vapor deposition, physical vapor deposition, laser techniques, and chemical methods [24]. Wide ranging applications of carbon nanotube has been observed in the field ranging from energy and electronics to automotive and space sectors [25–27]. Carbon nanotube has been used as a remarkable nanofiller to form the polymeric nanocomposites [28]. These nanomaterials have been recognized for number of superior structural/physical characteristics [29]. Small amounts of carbon nanotube have resulted in imperative properties of the nanocomposites. Furthermore, increasing nanofiller amounts have been found to increase the nanocomposite characters. To improve the effectiveness of carbon nanotube, functional nanofiller has been often included in the matrices. Difference between nanoparticles and nanocomposites can be seen as nanoparticles possess tens/hundreds of atoms of various shape/chemistry, whereas nanocomposite have nanolattices as part a part of the bulk matrix material.

Electron conduction and ionic conduction have been observed due to high aspect ratio of the nanotube and alignment in the matrices [30]. Carbon nanotube has been recognized for the formation of interconnecting network in the matrices leading to tunneling effect and so the effective electrical conductivity [31]. Owing to conductivity properties, carbon nanotube nanocomposites have been found functional for electrostatic/conductive coatings, electronic devices, textiles, transportation, engineering structures, and so on [32–34]. Another important application of carbon nanotube has been observed for sensing or detection of gaseous molecules, ions, and chemical species [35]. Consequently, carbon nanotube based nanomaterials have been employed for environmental relevance [36]. It has been observed that the sensing properties of carbon nanotube depend upon the charge or electron transport features [37–39]. Selectivity, sensitivity, and response times of carbon nanotube derived nanomaterials have studied for the carbon nanotube based sensors [40]. For sensors based on carbon nanotube nanocomposites, uniform dispersion of nanotube inn matrices, interface formation and choice of facile processing technique have been found indispensable. Among fabrication tactics, in situ, electropolymerization, coating, dipping, and solution methods have been mostly adopted for the formation of sensors [41–44]. For the analyte sensing, molecular interactions with the nanocomposite surface and interfaces, adsorption, and binding interactions have been investigated [45–47].

For carbon nanotube nanocomposite designs for sensors, conjugated polymers as well as nonconductive polymers like polyamide and olefinic polymers have been applied [48,49]. The resulting nanocomposites have effectively sensed the noxious gases and vapors. Akbari et al. [50] developed the carbon nanotube based field effect transistor as sensor for the ammonia analysis. Gas sensing mechanism was explained using a simple model and conductivity was analyzed through the current-voltage measurements. **Figure 1** shows the cylindrical tube like nanostructure of single walled carbon nanotube with hexagonally connected atoms.

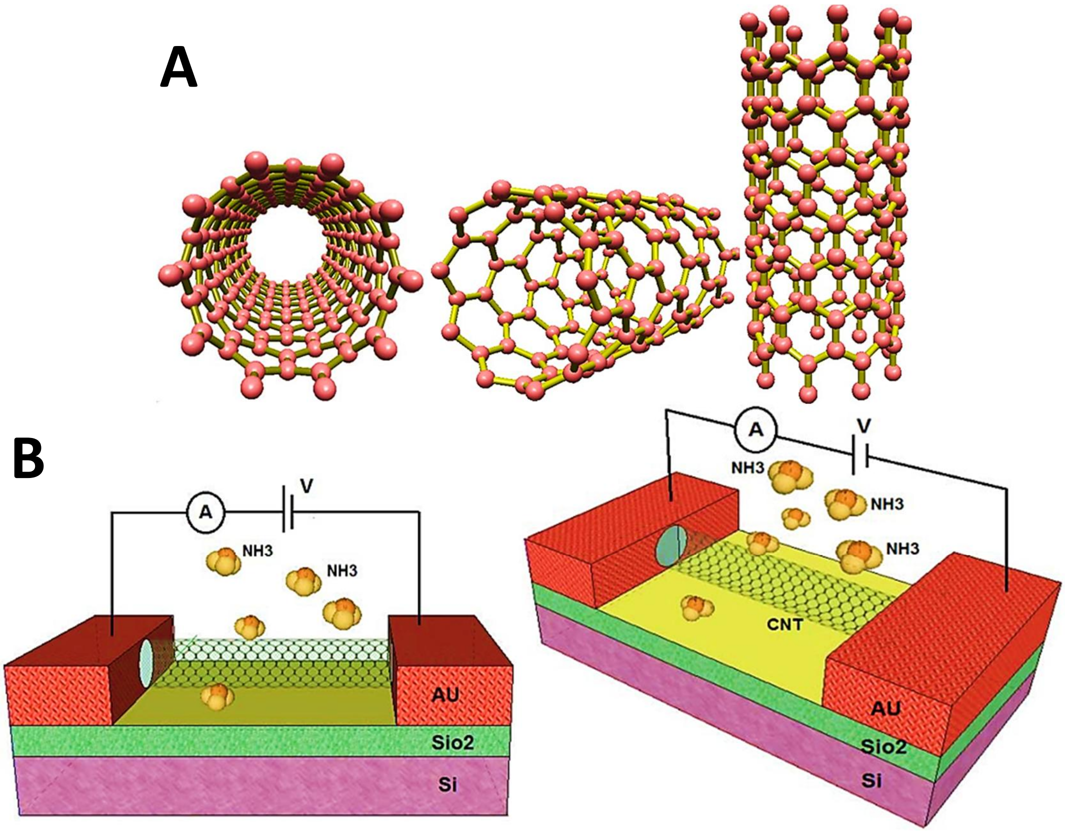

Figure 1. (A) Single walled carbon nanotube structures; and **(B)** Field effect transistor (FET)-based structure for a gas sensor with carbon nanotube channel [50]. Reproduced with permission from MDPI.

The length of nanotube has been found larger than the diameter to form a cylinder like nanostructure. The field effect transistor based gas sensor with carbon nanotube has also been presented. Conductivity responses of single walled carbon nanotube towards analyte gas molecules have been credited to the semiconducting nature of carbon nanotube. Like metal based field effect transistors, carbon nanotube have been observed to form conductive channels for the passage of electrons and detection of gaseous species. For the formation of carbon nanotube conducting channels, silicon and silica based layered dielectric substrate has been used. Upon the interaction of ammonia molecules with the field effect transistor, electron flow was observed in the external circuit for gas sensing analysis. Chiou et al. [51] designed the chemi-resistive gas sensor based on the poly(ethylene glycol)/multi walled carbon nanotube nanocomposite. The gas sensor was used to sense the acetone vapors at moderate temperature without using heat treatments. The sensing mechanism of the nanomaterial was also analyzed. **Figure 2** designates the testing apparatus used for the gas sensor. Here, mass flow controller was used to pass the acetone vapors over the sensors under controlled temperature and concentration. Multi steps were involved in the repeated test cycles during gas sensing. According to scanning electron microscopy, nanocomposite sensor had thickness of 217.6 nm (**Figure 3**). The nanotubes were observed enfolded with the polymer. The sensor response was also studied as a function temperature in the acetone concentration of 300–1000 ppm. The linear correlation coefficient was found around 0.98 at room temperature, as per fitting curves of sensing responses.

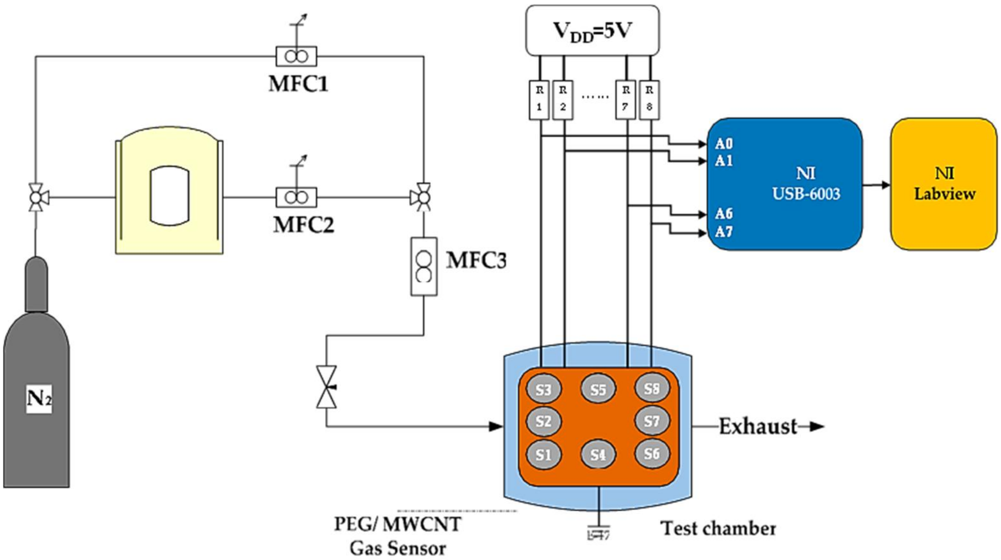

Figure 2. Test instrument of poly(ethylene glycol)/multi-walled carbon nanotube (PEG/MWCNT) gas sensor [51]. Reproduced with permission from MDPI.

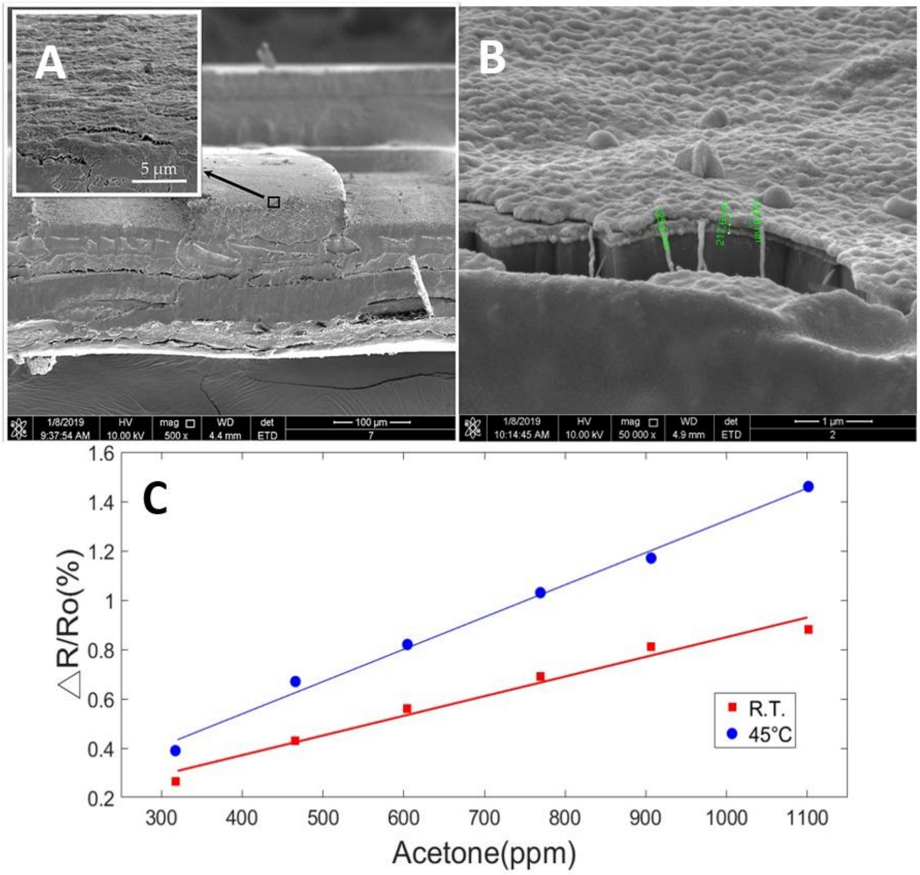

Figure 3. Scanning electron microscopy images of **(A)** close view of the interface between electrode and poly(ethylene glycol)/multi-walled carbon nanotube (PEG/MWCNT) nanocomposite film; **(B)** close view of PEG/MWCNT nanocomposite film; and **(C)** The fitting curves of sensor response at different operating temperature as a function of acetone concentration (300–1000 ppm) [51]. Reproduced with permission from MDPI.

Lapointe et al. [52] fabricated the carbon nanotube filled nylon 69 based nanomaterial for gas sensor. According to scanning electron microscopy micrograph, carbon nanotubes were found to be uniformly dispersed in the matrix (**Figure 4**). The fine dispersion and network formation led to the development of interfaces for gas interaction and analysis. On the field effect transistor, carbon nanotube was seemed to be dispersed and due to high surface area and interfaces, gaseous species were interacted and sensed.

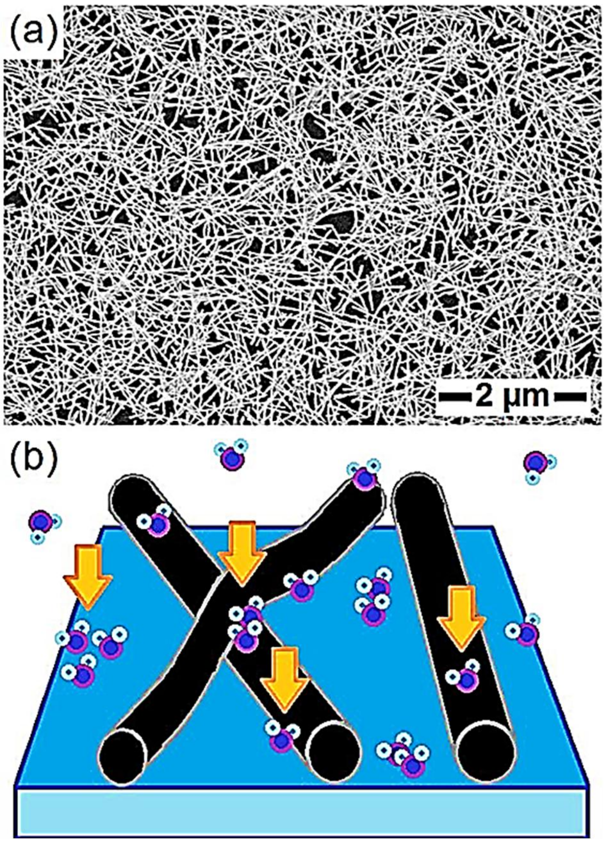

Figure 4. A percolation network of carbon nanotubes: **(a)** scanning electron microscopy of single walled carbon nanotubes on nylon 69; and **(b)** schematic illustration of the air solid interfaces where analytes may interact with the carbon nanotube network field effect transistors as highlighted by the arrows [52]. Reproduced with permission from ACS.

Figure 5 shows the scheme of carbon nanotube based field effect transistor having silica dielectric gate i.e., 1000 nm thick. The measurement of transfer characters of sensor revealed superior gas selectivity due to high aspect ratio of carbon nanotube. Using the trans-conductivity and threshold voltage, superior sensitivity and selectivity have been observed towards alcohol and organic molecules.

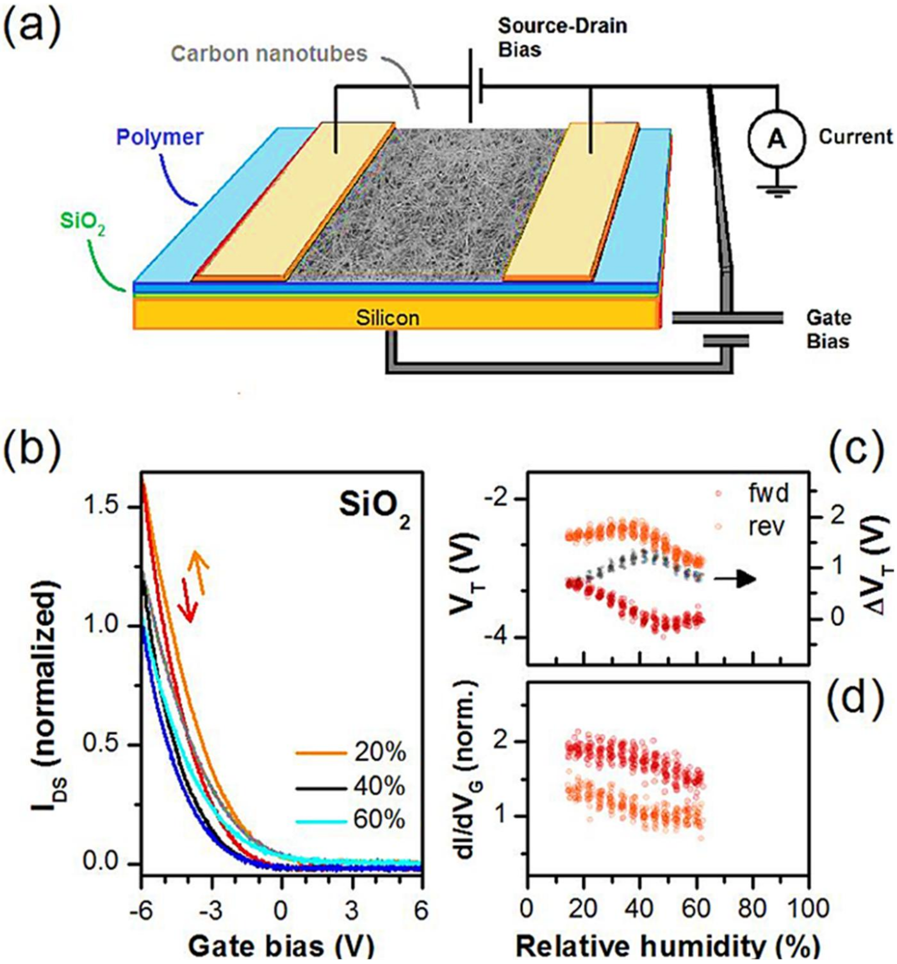

Figure 5. (a) Carbon nanotube network field effect transistor (CNN-FET) in bottom gate; and configuration as used in this work; **(b)** transfer characteristics of a CNN-FET with silica SiO_2 gate dielectric at normalized relative humidity of 60% RH; and **(c,d)** RH dependence of CNN-FET threshold voltage and transconductance determined from linear fit of transfer curves using the −6 V to −4 V range. Red and orange symbols correspond to forward (fwd) and reverse (rev) sweep directions, respectively, and black symbols show hysteresis between reverse and forward sweeps [52]. Reproduced with permission from ACS.

Among conjugated polymers, polyaniline, polypyrrole, and polythiophene have been applied for gas sensing [53]. The gaseous molecules of NO_2, SO_2, CO_2, methane, halogens, and other organic vapors have been sensed using the conjugated polymers [54–56]. Srivastava and co-workers [57] formed the single walled and multi walled carbon nanotube filled polyaniline nanocomposites through solution casting and spin coating. Change in resistance was used to assess the sensor response for hydrogen gas. High sensitivity was observed due to high surface area of carbon nanotube. Karmakar and colleagues [58] used polyaniline and carbon nanotube derived the sensing nanomaterial for sensing the NO_2 and CO_2 molecules. Miah and researchers [59] proposed a gas sensor based on polypyrrole/carbon nanotube nanocomposite for sensing NOx molecules. The nanocomposite was formed using the in situ method. Vijeth and workers [60] established the polythiophene and carbon nanotube derived nanocomposite using in situ oxidative method. The polythiophene/carbon nanotube based gas sensor was applied for hydrazine gas. The sensor had detection limit of 0.18 μM and sensitivity of 0.285 μA $\mu M^{-1} cm^{-2}$. Badhulika and co-workers [61] formed the single walled carbon nanotube filled poly(3,4-ethylenedioxythiophene):polystyrene nanomaterial for sensing of vapors like ethanol, methanol, and methyl ethyl ketone. Electropolymerization was used to form the nanocomposite. The resulting sensor response was recorded through the change in resistance values with enhancing vapor concentrations from 2.5% to 5%. Sensing mechanism was also studied using electrostatic effects. Sharma and researchers [62] developed the multi walled carbon nanotube filled poly(3,4-ethylenedioxythiophene):polystyrene nanocomposite sensor using solution method. The sensor was applied for sensing the ammonia gas. The sensitivity of 16% was attained with the response time of 15 min. The sensitivity of 5.59% was observed. In this manner, effective gas sensors have been fabricated using the carbon nanotube derived nanocomposites. **Table 1** demonstrates various convenient designs of carbon nanotube nanocomposites for gas sensing.

Table 1. Essential polymer/carbon nanotube nanocomposite in gas sensing.

Polymer/conjugated polymer	Nanofiller	Processing	Property/Application	Ref.
Nylon 69	Carbon nanotube	Solution method	Field effect transistor; solid interfaces; lock-and-key sensing mechanism; alcohol/organic solvent vapor sensing	[52]
Polyaniline	Carbon nanotube, zinc oxide nanorods	In situ oxidative polymerization technique	NOx and COx molecules; chemiresistive response 70 % at 120 ppm; recovery time <120 s	[58]
Polyaniline	Single walled carbon nanotube	Solution method	H_2 gas sensing response R_g/R_0 1.83	[57]
Polyaniline	Multi walled carbon nanotube	Solution method	H_2 gas sensing response R_g/R_0 2.30	[57]
Polyaniline	Carbon nanotube	Interfacial technique	Ammonia sensing	[63]
Polypyrrole	Carbon nanotube	Spin coating	Ammonia sensing	[64]
Polypyrrole	Carbon nanotube	In situ; spin coating methods	Ammonia sensor	[65]
Poly(3,4-ethylenedioxythiophene): Polystyrene	Multi walled carbon nanotube	Solution casting technique	NH_3 gas sensing; response time < 15 min; sensitivity 16%	[62]
Poly(3,4-ethylenedioxythiophene): Polystyrene	Single walled carbon nanotube	Electropolymerization	Volatile organic vapors methanol, ethanol, methyl ethyl ketone; ethanol and methyl ethyl ketone vapor detection; detection limit 5.95% and 3%, respectively	[61]

3. Graphene nanomaterials for gas sensing

Graphene has been categorized as an exclusive carbon nanomaterials having one atom thick nanosheet of carbon atoms [66]. Initially, graphene was discovered in 2010 [67]. Sp^2 hybridization and delocalization have been observed in the carbon atoms. Specific graphene nanostructure has led to the advanced structural and physical characteristic of this unique carbon nanomaterial. Most importantly, graphene has verry high Young's modulus of about I TPa, electron mobility of 200,000 $cm^2V^{-1}s^{-1}$, and thermal conductivity of 3000–5000 W/mK [68]. Graphene has been further modified in various forms to attain further high structural features. Effective methods for graphene formation involve graphite liquid or mechanical exfoliation, chemical vapor deposition, plasma and laser techniques, and organic synthesis approaches [69,70]. Essential modified forms of graphene have been recognized as graphene oxide (oxidized graphene nanosheet), reduced graphene oxide, graphene nanosheet with functional groups, and nanoparticle functional graphene nanosheets [71]. Among these, graphene oxide has been the most widely used functional form of graphene, commonly prepared through facile Hummer's method [72]. Applications of graphene has been found wide ranging from the electronics and engineering sectors to biomedical fields [73].

In environmental gas sensing application, carbon nanomaterials have gained increasing research curiosity owing to fine electrical conductivity and ionic or molecular sensitivity features. Graphene, with delocalized nanostructure, may form conjugated system with the conducting matrices to further enhance the conduction properties and resulting applications. Such systems have been studied for superior selectivity and sensitivity towards various gaseous species such as the oxides of nitrogen, sulfur, carbon, and other vapors. Consequently, conductive polymeric matrices have been used for gas sensing, however, several non-conjugated matrices or polymers have also been applied to for the effective gas sensors [74]. Noticeable conjugated polymers may include the polyaniline, polypyrrole, and polythiophene derivatives for gaseous detection. These polymers with graphene nanocarbon have been applied for the sensing of the gases like methane, halogen gases, and various noxious oxides of sulfur, nitrogen, or carbon [75]. For example, polyaniline and polypyrrole nanomaterials have been used to sense the NO_2 and CO_2 gaseous species [76,77]. Polythiophene and derivatives have been found to detect the hydrazine and toxic gases [78].

Pristine graphene has been applied as an important material for gas sensing [79,80]. With conjugated polymers, graphene has revealed further improved sensing behavior [81–83]. Graphene based sensors depicted visible change in resistance upon coming in contact with the analyte molecules [84]. For ion and gas sensing, combinations of polyaniline and graphene have exposed superior performance [85–87]. Wei et al. [88] reported on the long-period fiber grating based on graphene and formed surface plasmon resonance. **Figure 6** shows the design of graphene long period fiber grating/surface plasmon resonance and interaction with methane molecules. Using the CO_2 laser, nanomaterial was coated on the fiber core and then graphene long period fiber grating/surface plasmon resonance was coated on the silver film on silica substrate. Afterwards, graphene was deposited using the chemical vapor deposition method to form the final sensor design. **Figure 7** demonstrates that the sensor had resonance wavelength at 1541.3 nm, as per signal collected every 5 s. The response time was observed as 50 s. The 90% methane sensitivity was attained within 65 s. With the methane gas exposure, the resonance wavelength revealed increase/decrease behaviors. It was observed that the resonant wavelength observed was 0.05 nm for graphene long period fiber grating/surface plasmon resonance.

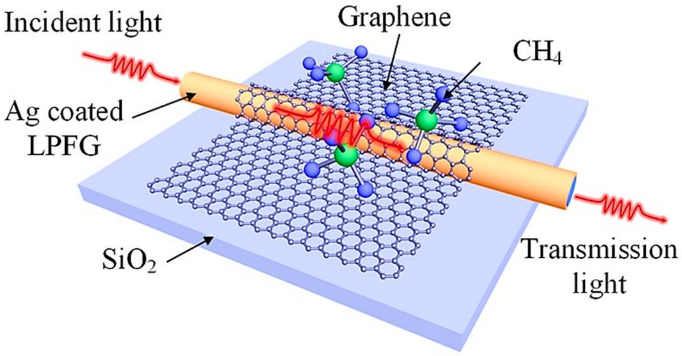

Figure 6. Schematic of graphene-based LPFG SPR sensor [88]. LPFG/SPR = long-period fiber grating/surface plasmon resonance. Reproduced with permission from MDPI.

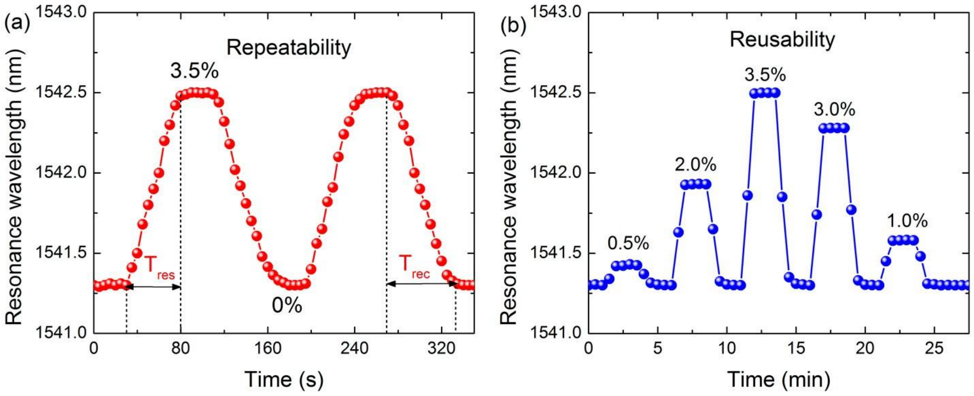

Figure 7. (a) Repeatability response curve of graphene-based LPFG SPR sensor to 3.5% methane gas sample; and **(b)** Reusability of graphene-based LPFG SPR sensor to methane gas sample with different concentrations [88]. LPFG/SPR = long-period fiber grating/surface plasmon resonance. Reproduced with permission from MDPI.

Wu et al. [89] fabricated the nanocomposite of polypropylene, polyaniline, and graphene based gas sensor. The nanomaterials have been formed using the in situ method and dip coating process. The resulting polypropylene/graphene/polyaniline nanocomposite derived gas sensor was used for ammonia gas sensing. The nanocomposite has fine interconnected hierarchical nanostructure for fine molecular analysis. **Figure 8** shows the sensing mechanism of the nanocomposite depending upon the doping and de-doping processes and charge transportation occurring at interfacial areas [90]. The gas senor was used for noxious gases sensing in exhaled human breath.

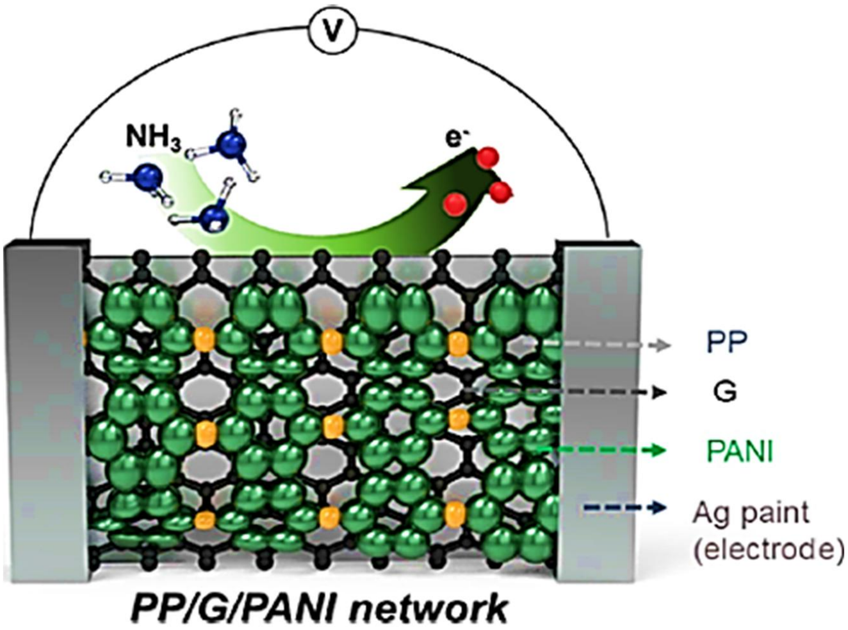

Figure 8. Sensing mechanism of PP/G/PANI hybrid sensors [89]. PP = polypropylene; PANI = polyaniline; G = graphene; PP/G/PANI = polypropylene/graphene/polyaniline. Reproduced with permission from ACS.

Consequently, volatile sulfur compound like H_2S gas (2%) and ammonia gas (100 ppb in 114 s) were effectively sensed in the exhaled human breath (**Figure 9**). As compared with the pristine polyaniline, the polypropylene/graphene/polyaniline nanocomposite sensor had 250% superior sensing performance [91].

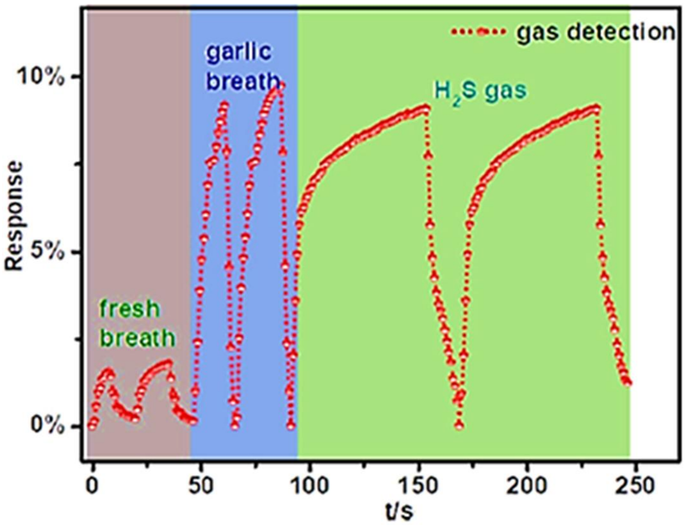

Figure 9. Photograph of PP/G/PANI sensor for volatile sulfur compounds in human breath and pure H_2S [89]. PP/G/PANI = polypropylene/graphene/polyaniline; Reproduced with permission from ACS.

Tange et al. [92] fabricated the polyaniline and graphene oxide derived nanocomposites through in situ polymerization. The microstructure, electrochemical, and electrical conductivity properties have been studied for the nanocomposites. The nanocomposites have fine nanofiller dispersion and interactions between the matrix and nanofiller. For structural analysis, X Ray diffraction patterns of polyaniline, graphene oxide, polyaniline/graphene oxide, polyaniline/modified graphene oxide were scanned (**Figure 10**). Neat polyaniline had crystalline peaks appeared at 2θ of 19.4° and 25.4°, while graphene oxide depicted peak at 2θ of 10.12° due to (001). In modified graphene oxide, the shift in diffraction peak was observed to 9.90° due to change in interlayer spacing. In the formation of nanocomposites with polyaniline, the diffraction pattern had amorphous nature and there was decrease in the crystalline order. Consequently, the electrochemical characters of the nanocomposites were enhanced. In addition, electrical conductivity of the nanocomposite was increased to 508 Sm^{-1} was attained.

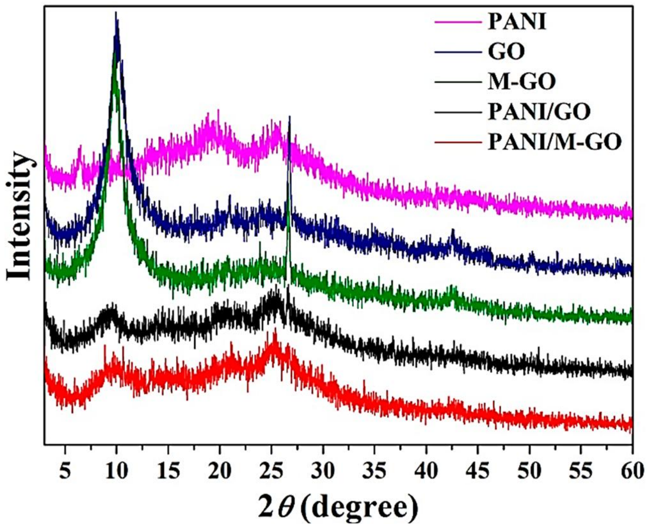

Figure 10. X-ray diffraction patterns of PANI, GO, M-GO, PANI/GO, and PANI/M-GO nanocomposite [92]. PANI = Polyaniline; GO = graphene oxide; PANI/GO = polyaniline/graphene oxide; PANI/M-GO = polyaniline/modified graphene oxide. Reproduced with permission from MDPI.

Similarly, effective combinations of conjugated polymers with graphene, graphene oxide, or reduced graphene oxide have been developed for the sensing of gaseous species like methane, methanol, hydrogen, ammonia, and so on [93,94]. **Table 2** illustrates the gas sensitivity behavior of various combinations of conjugated polymers and graphene nanofillers. In such systems, polymers and graphene nanofillers have been found to develop a charge transfer complex and electron donor-acceptor nanostructures for sensing of analyte gases.

By comparing the nanocarbon based gas sensors with inorganic nanoparticles filled sensors, the efficiency can be analyzed. For example, Bonyani et al. [95] reported on the gold decorated zinc oxide nanoparticle based polyaniline nanocomposite sensors. The 9 nm zinc oxide nanoparticles were prepared and filled in 10–50 wt.% in the polyaniline matrix and gas sensing response was analyzed. Fine gas sensor response was analyzed at 300 °C for NO_2 gas. The $\Delta R/R_0$ sensor response was found comparable to the polyaniline/graphene nanocomposite sensor. Bairi et al. [96] formed tanninsulfonic acid doped polyaniline and titania nanocomposites for ammonia gas sensor using in situ and spin coating. The gas sensor was tested in the ammonia concentration range of 20–60 ppm. The sensor had revealed 90% change in resistivity, which is higher than the sensitivity of polyaniline/graphene nanocomposite observed [97]. The comparative analysis revealed that the inorganic nanoparticles filled conjugated polymer sensors may have higher performance than the graphene or carbon nanotube based sensors, depending upon the material design.

It has been observed that the crystal size or grain size of the nanofiller particles in polymeric nanocomposite played important role for gas sensing performance. By reducing the crystal or particle sizes, the sensitivity as well as response speed of the sensors were considerably enhanced [98]. Decrease in sizes usually causes increase in surface area of the nanoparticles, which is used to enhance the vacancies by decreasing the free electrons concentration. Consequently, the adsorption of gas molecules is improved, in turn enhancing the gas sensing response.

Table 2. Provisions of polymer/graphene nanocomposites in gas sensing.

Conjugated polymer	**Nanofiller**	**Processing**	**Property/Application**	**Ref**
Long-period fiber grating	Graphene	Coating	Plasmon resonance sensor; sensitivity CH_4 1%–3%	[88]
Polypropylene/polyaniline	Graphene	In situ polymerization; dip coating	H_2S gas; detection limit 100 ppb; NH_3 gas sensing; response time 114 s	[89]
Polyaniline	Graphene	Interfacial technique	H_2O_2 sensing	[99]
Polyaniline	Graphene oxide	In situ method	Methanol sensitivity; electrical conductivity 241 Sm^{-1}	[100]
Polyaniline	Graphene	Layer-by-layer technique	π-π conjugation; high methane sensitivity	[101]
Polythiophene	Reduced graphene oxide	In situ method	Humidity sensor	[102]
Polyaniline/Palladium	Reduced graphene oxide	Deposition technique	H_2 gas; $\Delta R/R_0 = 25\%$; Response time 20 s	[103]
Polyaniline	Reduced graphene oxide	In situ method	NH_3 sensing; response 59.2% at 50 ppm; response time 20 s	[97]
Polyaniline	Graphene	In situ method	$\Delta R/R_0 = 30\%$;	[104]
Polypyrene	Graphene oxide	Electrochemical co-deposition	Linear reversible response; sensitivity 9.87×10^{-4}	[105]
Poly(methyl methacrylate)	Graphene	Solution method	Octanoic acid detection; current response per power law with exponent in the range 0.4–0.8	[106]

4. Prospects and future scenarios

Fabrication of the gas sensing nanocomposites with graphene or carbon nanotube has been mostly carried out using the solution and in situ route. The solution mixing method has been most commonly applied for the formation of nanocarbon nanocomposites owing to low price and facile parameters [107]. This technique includes the simple mixing of polymer and nanoparticles in a suitable solvent and then the material casting through solvent evaporation. The technique has been found practicable for variety of thermoplastic and conjugated matrices [108]. In the presence of a suitable solvent, the polymers or monomers are adsorbed on the nanocarbon nanoparticles to enhance the interactions and dispersion [109,110]. Consequently, the solution formed nanocomposites have revealed desired physical and technical features [111]. In situ polymerization has also been extensively used to form the carbon nanocomposites for sensors. In this technique, first the monomer molecules are dispersed in a solvent and then subsequently polymerized with the pre dispersed nanoparticles [112]. Monomers are finely adsorbed on the nanocarbon nanoparticles for better dispersion and nanocomposite production [113]. Both the solution casting and in situ technique possess advantages of using non-toxic solvents and environmental friendliness. These methods have been known for well-matched interface formation and compatibility of the nanocomposites [114].

Consequently, high-tech carbon nanotube nanocomposites and graphene nanocomposites have been reported to be utilized in the gas sensing applications. Nanoparticle type, contents, interaction with matrices, dispersion, and interfacial effects influence the sensor performance [115]. Consequently, carbon nanoparticles like carbon nanotube or graphene may form interconnecting network in the matrices for electron conduction, percolation effects, and development of charge transfer complex or π-π interactions leading to fine sensing performance [116]. In addition, nanocarbon nanocomposites have advantages of robustness, functioning reliability, and environmental stability features [117]. Better nanoparticle dispersion has been analyzed as an important feature to enhance the electrical conduction and sensing performance [118]. Conversely, nanoparticle aggregation or self-association may deteriorate the sensing and conductivity performance of the nanocomposites. Besides the nanoparticle nanofiller, type of polymer or matrix has been found important to define the sensing behavior of nanocomposite. Various nanocarbon based designs have been formed and utilized for sensing the gaseous molecules like hydrogen, hydrocarbon, ammonia, carbon dioxide, halogens, and range of toxic oxides. Bulk of literature is available on the conjugated polymer with carbon nanoparticles for sensing designs, however non-conjugated systems have been less explored for gas sensing applications. Despite of the research so far, there is need of new sensing designs to be explored for future high performance nanomaterials [119]. There is need of focused research labors to find out the real sensing mechanism for the gas detection and possibilities for the development of next-generation sensors [120]. Facile methods like in situ, solution, and coating have been applied to form the gas sensors, still there is need of applying sophisticated techniques like three or four dimensional printing to form more precise nanomaterial designs [121]. Printing techniques can be beneficial for not only designing the sensors with optimized structure and conditions but also the superior sensitivity, selectivity, and responsiveness of the nanomaterials due to better compatibility, material interactions, and synergistic effects. Further exertions on the fabrication of modified nanocarbon based sensors may lead to marvelous high performance sensor designs.

5. Conclusions

Concisely, this overview hearsays competent carbon nanotube and graphene based nanocomposite designs utilized for gas sensors. Consequently, the nanomaterials have been investigated for microstructure, and structural as well as physical properties. Specific features regarding the gas sensing such as conductivity, change in resistance, nanomaterial sensitivity, selectivity, detection limit, response time, etc. have been explored to analyze the sensor performance. It has been observed that the carbon nanostructures like graphene and carbon nanotube have been efficiently adopted for gas sensor designs. The nanocarbon nanostructures have been mostly used in the nanocomposite form with polymer matrices. Thermoplastic and conductive polymers have been filled with the nanocarbon nanoparticles to form the desired sensing nanomaterials. Among these sensors, polyaniline and nanocarbon based gas sensors may have response time of 20–100 s. These sensors can have response of up to 60% and $\Delta R/R_0$ was detected in the range of 20%–30%. Fine dispersion of nanocarbon nanoparticles and network formation in matrices have led to superior electron conduction and so the sensing properties of the nanocomposites. The resulting gas sensors have been found to analyze various noxious gas species like oxides of nitrogen, carbon, or sulfur, halogens, hydrocarbon or organic vapors, and others. Although, significant literature has been found in this regard, nevertheless there is need of further research efforts in this direction for better nanomaterial design and property optimization and exploration of the sensing mechanisms involved. Though, research up till now has anticipated abundant designs for gas sensors, as per studies development of new graphene and carbon nanotube based sensors may disclose technical fields due to advantageous performance. Using functional carbon nanotube and graphene based sensors may reveal low price, high electrical conductivity, electrochemical performance, stability, sensitivity, and reproducibility. New gas sensing designs can be advantageous for potential environment, energy, beverages, and medical industries. The desired gas molecules can be easily detected for uses in these sectors.

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