Air Pollution Monitoring Using Near Room Temperature Resistive Gas Sensors: A Review

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Abstract

The growth of industries and other human activities have led to ever increasing amounts of pollutants in both outdoor and indoor spaces. These pollutants have hazardous effects on humans and the wider ecology. Hence, air quality monitoring (AQM) is essential and involves the robust monitoring of various toxic gases and volatile organic compounds (VOCs) - in case the concentration of any pollutant exceeds the safe limit in a given location. This paper describes the different sources of indoor and outdoor pollutants, reviews the current status of gas sensors, and discusses the role of new two-dimensional (2D) materials in detecting these hazardous gases at low power, i.e. close to ambient temperature. Here we review different synthesis techniques of 2D materials and discuss the sensing performances of pristine and functionalized nanomaterials for some of the important pollutants like NO_x, NH_3, SO_x, CO, formaldehyde and toluene. The review concludes with some proposed methods to help reduce air pollution today.

Keywords: Air quality monitoring, Resistive gas sensors, 2D nanomaterials, Near room temperature sensing

1. Introduction

There has been increasing interest to develop new low cost, low power gas sensors for various application-specific areas. This includes air pollution monitoring of both indoor and outdoor spaces, detection of toxic gases in and near industrial premises and also sensors for biomedical applications [1]. There has been a rapid rise in levels of toxic gases and volatile organic compounds (VOCs) in air in recent years; particularly in urban spaces. This is mostly true for many cities in under-developed or developing countries. For example, a recent report (2018) from World Health Organization (WHO) shows that fifteen Indian cities and twenty-one Chinese cities are amongst the fifty most polluted cities in the world [2]. In 2019, air pollution is considered WHO as the greatest environmental risk to health [3]. The major sources of polluted air are fuelwood and biomass burning, burning of agricultural wastes (large scale crop residue burning takes place during winter), fuel adulteration, uncontrolled emission from vehicles and factories, traffic congestion and rapid construction [4]. These cause smog and hence increase airborne particulate matter (e.g. PM_{10}, $PM_{2.5}$), NO_x, NH_3, SO_x, CO and other VOCs in the air. All these pollutants are well in excess of human permissible limit in capital cities like Delhi (capital of India, 6th in WHO list), Kampala (capital of Uganda, 16th in WHO list), Doha (capital of Qatar, 21st in WHO list), Kabul (capital of Afghanistan, 28th in WHO list) and so on [2]. The situation is much better in developed countries because of better awareness and stricter regulations. Quality of indoor air is of equal importance as that of outdoor air, because we spend most of our time in indoor areas and it can have many times the level of outside pollution! Smart buildings (e.g. houses, hospitals, schools at smart cities in developed countries) are such places where different hazardous gases and VOCs like CO_2, CO, formaldehyde, benzene, toluene, ethylbenzene and xylene (popularly known as BTEX) along with humidity are monitored and restricted to permissible limit through proper detection and ventilation [5]. However, there is a lack of awareness and a detailed understanding about the long-term acute effect of these air pollutants among populations. Excessive exposure of air pollutants leads to increasing respiratory and cardiovascular diseases, such as acute lower respiratory infections (ALRI), chronic obstructive pulmonary disease (COPD), lung cancer, ischemic heart disease (IHD), pneumonia, and strokes [6, 7]. Thus, air pollution is the reason behind many diseases that often prove to be lethal. Almost 7 million deaths were caused globally by household and outdoor pollution in 2016 [2], the fourth-highest cause of deaths worldwide. Thus, the effect of air pollution is alarmingly high and much worse than many of us anticipate. This makes air quality monitoring (AQM) urgent and essential.

There have been many review papers published on metal oxide based resistive sensors for detecting toxic gases over the years [8-10]. There are also many reports on air pollution [4, 11]. Recently, several review papers highlighted two-dimensional (2D) layered material based resistive sensors [12-15]. However, the authors believe it is necessary and of utmost important to review: sources of air pollution; resistive sensors available in the market for detecting toxic gases; highlighting the necessity to develop near room temperature resistive sensors (which will reduce the power consumption drastically and fetch the way for '*things*' in the Internet); and possible approaches to tackle with air pollution.

2. **Sources of Air Pollutants**:

The rapid industrialization across various parts of Asia (particularly in India and China) in recent years has been an economic boon. Unfortunately, industrialization and rising the standard of living are coupled with poor implementation of governmental rules and regulations, which result in increased concentration of air pollutants. Fig. 1 shows the different indoor and outdoor pollutants that are generally present in the air because of different human activities. The pollutants shown in Fig. 1 are harmful to living beings and the environment if exposed beyond permissible exposure limits (PEL) as published by occupational safety and health administration (OSHA), United States [16, 17]. The Environmental Protection Agency (EPA), United States has also set National Ambient Air Quality Standards (NAAQS) for particulate matters $PM_{2.5}$ and PM_{10} [18]. Public agencies with similar spirit and purpose exist across the globe and set similar standards for the air pollutants. In order to devise a mechanism to monitor and check air pollution, it is necessary to

develop gas sensors that can detect these gases/VOCs. Current trends in gas sensors are presented in the next section.

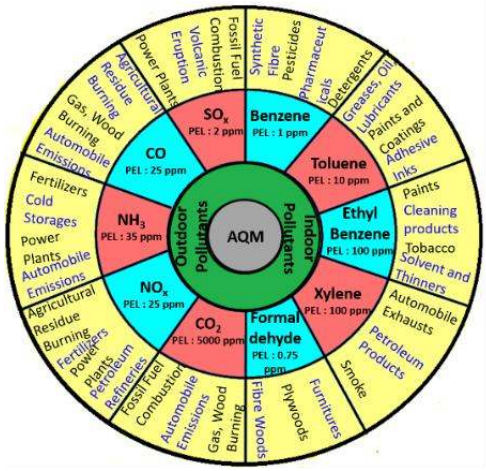

Fig. 1 Different indoor and outdoor pollutants with their safe-limits and sources

3. **Current Gas Sensors**, *motivations for near room temperature sensors*:

Any device that exhibits change in one of its fundamental properties like mass, capacitance, resistance etc. when placed in the proximity of a gas species can be classified as a gas sensor. Depending on the transduction methods, they can be categorized into electrochemical sensors, optical sensors, SAW based gas sensors, calorimetric sensors, chemoresistive sensors etc. Current commercial gas sensors are mostly based on electrochemical transduction. Such sensors are usually fabricated in a semi-automated batch process and it is not possible to integrate these with established micro-electronics technology, hence they are expensive [19]. A typical electrochemical sensor costs around $20, with evaluation board a total sensor system might cost well over $100 [20]. Other popular gas sensors, e.g. infrared gas sensor, is selective but much more expensive (>$1000). Most of the Indoor Air Quality (IAQ) systems sold in the market do not have provision to show the concentrations of individual VOCs, rather they measure total VOC content (TVOC) [21]. Also, they are very expensive, because most of the units are a combination of discrete sensors with printed circuit board (PCB) level electronics for integration along with data-analytics for pattern recognition. The cost of HERACLES Neo Electronic Nose which is manufactured and sold by renowned electronic nose manufacturer (Alpha MOS) is more than 100,000 euro [3]. The device, in its present form, also requires a skilled person to operating it.

Micro-resistive sensors have a clear edge over other sensors (electrochemical, IR, photoionization detector etc.), because the former is easier to integrate with conventional microelectronics technology. This makes the sensor devices more reliable (automated fabrication process) and much lower cost (for volume production) [22]. The heart of such a gas sensor is the sensing layer. Metal oxides (e.g. *n*-type SnO_2, ZnO, WO_3 and *p*-type NiO) are the gold standard materials that are commonly used as sensing layers for such resistive sensors [23]. Metal oxides are semiconducting in nature and interact with chemical analytes at elevated temperatures (150–400°C). At such high temperatures, oxygen is adsorbed at the surface of the metal oxide clusters by trapping electrons from the bulk. This increases (*n*-type material) or decreases (*p*-type material) the resistance of the sensing layer. The oxygen species (O^-, O^{2-} etc.) interact with the chemical analytes and release back electrons to the sensing layer. This change the resistance of the sensing layer, which is proportional to the concentration of the targeted chemical analytes. Fig. 2 is a schematic representation of the sensing mechanism of the metal oxides.

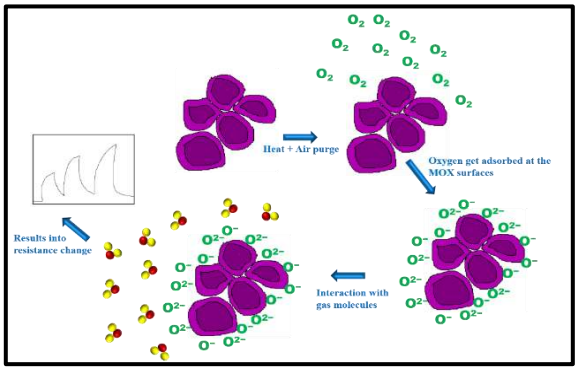

Fig. 2 Schematic representation of sensing mechanism of metal oxide nanoclusters with depletion/accumulation layers

Although metal oxides are highly sensitive, they are not selective. Also, they consume considerable power (~ 100-500 mW) for sensing target gases because of high temperature requirements [24]. This is not ideal for battery operated devices. Moreover, high sensing temperatures can affect circuit performance if the sensor is integrated on the same silicon die. In this respect, considerable breakthrough has been made by implementing sensors in innovative micro-hotplates (CMOS-MEMS integration), where heat will be confined in the membrane region (make the *sensor/circuit* integration possible), and also power consumption can be reduced because of low thermal mass and air insulation [25-28]. The micro-hotplates can be realized by front etching (suspended membrane) or back etching (closed membrane) techniques [20, 29-31]. A typical micro-hotplate contains a micro-heater (to heat up the sensing layer), interdigitated electrodes (to measure the resistance of sensing layer) under a nano-material based sensing layer (as shown in Fig. 3). The sensing layer is realized using post CMOS process, which is very challenging because in-situ synthesis of nano-material often requires harsh chemicals and high temperature, which might damage the on-chip circuitry and also the membrane. In this respect processes like local growth [32], localized hydrothermal technique [26], inkjet printing [33], printing using dip pen nano lithography [30, 31] etc. were developed.

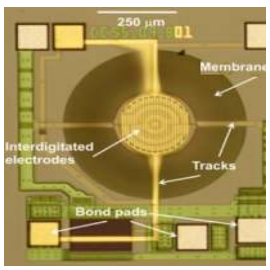

Fig. 3 Optical image of an interdigitated electrode integrated with micro-heater on micro-hotplate (adapted from [31])

Such resistive sensors are power efficient, for example, a temperature of 300°C can be achieved with few 10s of mW. However, the holy grail of resistive sensors can be achieved if the sensor operates at room temperature without deteriorating its performance. This will not only reduce power consumption almost completely, but also reduce the complexity associated with device fabrication. Thus, there is a need to develop high-quality, low-cost, ultra-low power sensors for many applications including air quality monitoring. Recent researches show that two dimensional (2D) layered materials especially carbon nanomaterials and transition metal dichalcogenides (TMDs) have potential to sense gases at near room temperature, however response from such materials are often poorer than the conventional metal oxides [34, 35]. So, in the following sections we will concentrate on synthesis techniques of carbon nanomaterials and TMDs, study their sensing capabilities reported in the literature so far and ways to improve their sensitivity so that their performance even at room temperature can match with that of metal oxides which usually operate at higher temperatures.

4. Synthesis Techniques of pristine 2D layered nanomaterials

The synthesis techniques of a nanomaterial can be broadly classified into two categories: top-down approach and bottom-up approach. Different methods in each category have been explored over the years to customize the synthesis of 2D layered materials for different applications. For example, fabrication of transistors requires highly pure nanomaterials whereas for gas sensing, nanomaterials with inherent defects and functional groups are preferable. This review focuses on discussing some of the widely explored techniques which are frequently employed for the synthesis of these nanomaterials.

4.1 Mechanical exfoliation – This is the simplest and most economic technique for synthesis of nanomaterials. It is a top-down method which is employed to reduce mechanically one or more dimensions of bulk materials to nanoscale. In 2004, Novoselov *et al.* produced highly pure graphene flakes by isolating monolayer carbon sheets of highly oriented pyrolytic graphite (HOPG) using this method [36]. The method is renowned as the "Scotch-tape method". Later on, this technique was extended to obtain other layered materials [37-40].Though there are reports on fabricating gas sensors using layered materials synthesized by mechanical exfoliation [41], this method is not generally preferable because of lack of repeatability, poor scalability, and difficulty to introduce functional groups to sensing layer.

4.2 Chemical vapour deposition (CVD) – This bottom-up technique of nanomaterials synthesis is based on the chemical reactions that occur between the precursors and substrate thereby producing high quality 2D material thin films of large area. The molecules of the gas precursors are fed into a reactor and passed through a hot zone (700–1200°C) where the reactants dissociate and deposit onto the substrate, which is placed at a relatively lower temperature in the reactor [42]. There are different parameters which guide the dynamics of this growth process. For graphene, usually a mixture of any hydrocarbon and H_2 are used as precursors and transition metals, mostly Cu or Ni is used as the catalytic substrate [43]. For TMDs, it is a two-zone process. Zone-1 consists of precursors (MoO_2/MoO_3 for MoS_2 and WO_3 for WS_2) in powder form, which is heated to a temperature of 700–900°C; and Zone-2 consists of sulphur powder, which is heated to a temperature of 250°C. Then both the zones are cooled down after a certain time leading to the formation of triangular WS_2 or MoS_2 sheets [44-46]. The commonly preferred substrate for CVD growth of TMDs is SiO_2 coated Si [47]. However, efficient methods of transferring the CVD grown thin films from the metal or Si/SiO_2 surfaces to substrates like polymers have already been developed and are extensively used [48]. This had enabled development of flexible sensors that are rugged, wearable and tailorable. CVD is one of the widely used methods for fabricating defect-free layered materials to realize electronic devices, however its application in sensing is limited simply because of lack of defects. Also, CVD is one of the more expensive and sophisticated methods for production of large-area 2D materials.

4.3 Epitaxial Growth – This is another sophisticated method of synthesis of nanomaterials. For epitaxial growth of any nanomaterial it is necessary to have a substrate with specific crystallographic orientation so that the atoms of the substrate can coalesce into a uniform, single-crystal layer of that particular nanomaterial with a reduced number of grain boundaries [49]. One of the potential ways for producing large area graphene layers is epitaxial growth of graphene on SiC substrates [50]. In this technique, SiC substrate is heated to high temperature of around 1200°C under high vacuum (~10^{-6} Torr). Under such conditions, the silicon atoms evaporate from the substrate and the carbon atoms rearrange themselves to form graphene layers. Epitaxial growth of TMDs is mostly intertwined with CVD process and is still developing [51]. For WS_2, Polyakov *et al.* reported epitaxial growth of WS_2 (0001) layers on ZnO/ WO_3 core shell structure [52]. Again, epitaxially grown material is usually pure and not very useful for gas sensing, so the readers interested in this growth technique are advised to refer to the cited works for more details.

4.4 Liquid exfoliation – This is a low-cost technique for nanomaterial synthesis in which the bulk precursor is exfoliated using suitable chemical reagents. The method of isolating different nanosheets using liquid exfoliation is schematically represented in Fig. 4.

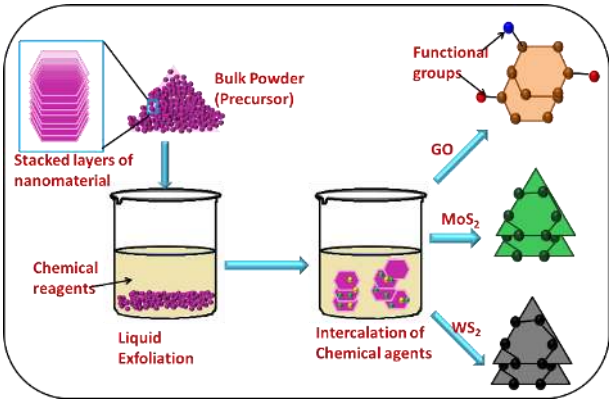

Fig. 4 Schematic (not to scale) demonstrating the steps of liquid exfoliation of different layered nanomaterials

The chemical agents intercalate between the layers of the bulk materials and isolate monolayers or few layers of nanosheets. In the process, some of chemical species get attached to edges or basal planes of the nanosheets and hence the purity of nanomaterials synthesized using this technique is not very high. Also, liquid exfoliation can cause defects, e.g. by creating sulphur vacancies in MoS_2 or WS_2 [53]. But for sensing applications this is an added advantage as the chemical species and defect sites act as

attaching/interacting sites for the gas molecules and this is why liquid exfoliation method is widely used to synthesize nanomaterials for gas sensing applications. Different methods and recipes have been explored to produce reduced graphene oxide (RGO), which is a derivative of graphene, using chemical routes. The first step of almost all methods is to synthesize graphene oxide (GO). This is done by rigorous chemical treatment of graphite powder that can be accomplished using the Hummers [54], Brodie [55] or Staudenmaier [56] method. Among the three ways to synthesize GO, Hummers method is the one that is most widely used. GO is insulating in nature and hence in order to get back its conductivity, GO has to be reduced. GO can be reduced chemically [57], thermally [57], electrochemically [58] or by photocatalytic reduction [59]. The extent of reduction of GO to RGO can be quantitatively determined by finding out their C/O ratios using X-ray photoelectron spectroscopy (XPS) as is shown in Fig. 5 (a-b). Although none of reduction method is efficient enough to produce reduce graphene oxide (RGO) nanosheets having properties exactly same as those of pristine graphene, but development of this wet chemical method enabled researchers to produce graphene (with controlled amounts of functional groups) in bulk at a very cost-effective level. For TMDs, wet synthesis does not require oxidizing the bulk material. Different chemical reagents that are being used for exfoliation, intercalate between the layers of the TMDs and thus isolate each layer. These intercalated species can then be filtered out by using centrifuge. But as the bonds between the TMD sheets are already broken by then, so the nanosheets remain isolated even after the purification is carried out. For example, Pagona *et al.* reported exfoliation of 2H-MoS_2 and 2H-WS_2 using chlorosulfonic acid [60]. There are several other reports on liquid/chemical exfoliation of TMDs [53, 61, 62]. Since the monolayer isolation in case of TMDs do not involve rigorous chemical procedure, so, it is possible to produce 2D layers of these materials without compromising their purity. XPS is also employed to assess the purity of single/few layered TMDs synthesized using liquid exfoliation as is shown in Fig. 5 (c-f).

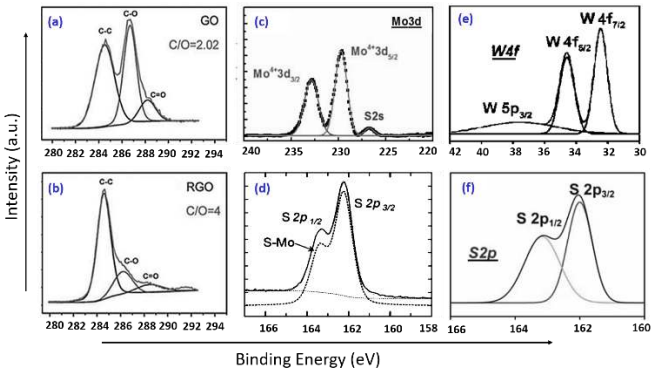

Fig. 5 XPS results of (a-b) GO and RGO with respective C/O ratio [63] (c-d) 2H-MoS_2 (e-f) 2H-WS_2 [64, 65] 2H-WS_2 [66]

Apart from XPS, Raman spectroscopy, Fourier transform infrared spectroscopy (FTIR), UV-Vis spectroscopy and X-ray diffraction (XRD) are also used to study the presence of different bonds and the crystal structures of these sensing materials. Sophisticated techniques like atomic force microscopy (AFM), transmission electron microscopy (TEM), scanning electron microscopy (SEM) are employed for the morphological characterizations of these nanomaterials. The AFM image of GO synthesized using Hummers method is shown in Fig. 6 (a). Fig. 6 (b) shows TEM image of triangular MoS_2 nanoflakes.

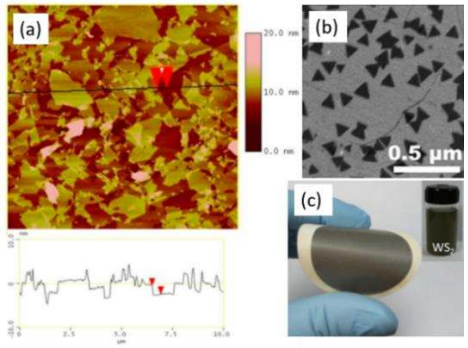

Fig. 6 (a) AFM image of GO produced using Hummers method (Reprinted with the permission from [67] Copyright©2013 American Chemical Society) (b) SEM image of MoS_2 nanoflakes [68] and (c) WS_2 layer deposited on PVDF (flexible) substrate [69]

In addition to the aforementioned advantages offered by liquid/chemical exfoliation techniques of synthesis of nanomaterials, these also allow the sensing materials to be deposited over any substrate including flexible platforms using drop casting, spin coating, dip coating or vacuum filtration method. Fig. 6 (c) shows WS_2 coated on flexible PVDF substrate. Once it is ensured that the 2D materials have been synthesized properly by the aids of different characterization techniques, these are used to fabricate the sensors and the devices thereafter are extensively tested for their sensing capabilities. The next section discusses the sensing performances exhibited by different 2D materials near room temperature.

5. Gas sensing by pristine 2D layered nanomaterials

Monitoring the presence of the pollutants requires development of efficient devices for the continuous detection of these gases and VOCs. 2D materials, owing to their outstanding properties can sense different analytes. This review however, would limit its focus on humidity and some of the important indoor and outdoor pollutants namely NO_x, NH_3, SO_x, CO, toluene and formaldehyde.

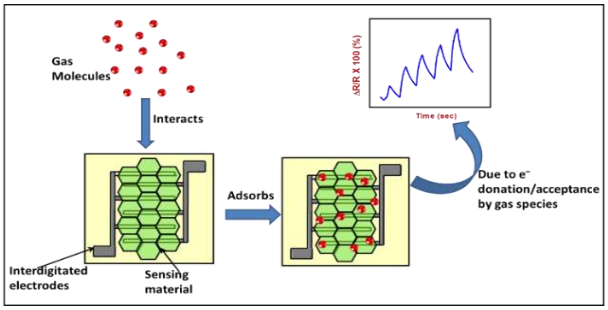

Fig. 7 Schematic representation of working of a resistive gas sensor

Fig. 7 schematically represents the working of a generic chemoresistive gas sensor, here sensor resistance changes when gas molecules get adsorbed on its surface. The response of sensor is calculated as $\frac{R_{gas}-R_{air}}{R_{air}} \times 100\% = \frac{\Delta R}{R_{air}} \times 100\%$

As the surface-to-volume ratio of the 2D nanomaterials is very high they offer high sensitivity towards the species in their proximity. The next section reviews the sensing performance of the intrinsic 2D materials.

5.1 Graphene based gas sensors: Graphene has outstanding electrical (conductivity and carrier mobilities) and mechanical properties which are desired for gas sensing [70]. Pristine graphene is primarily known for sensing NH_3 [71], NO_x [72], H_2O [73] and some VOCs [74]. The first article on graphene-based gas detectors was reported in 2007 by Schedin *et al.* in which they demonstrated that mechanically exfoliated graphene sheets have the potential to detect single molecules of gases [75]. Rivera *et al.* reported NH_3, NO_2 and ethanol sensing by graphene synthesized using CVD [76]. Studies on determining the sensing mechanism of graphene were also performed by different research groups and it was found that the gas molecules are physisorbed at the surface of the graphene. Chen *et al.* demonstrated the possible adsorption sites on a graphene sheet (Fig. 8(a)) [77]. The gas molecules coming in the proximity of graphene, occupy either of the sites as shown in Fig. 8(a) thereby changing the electrical conductivity of the sensor. This change in conductivity of the graphene layer is attributed to the change in the local carrier concentration induced by the surface adsorbates which act as either electron donors or acceptors.

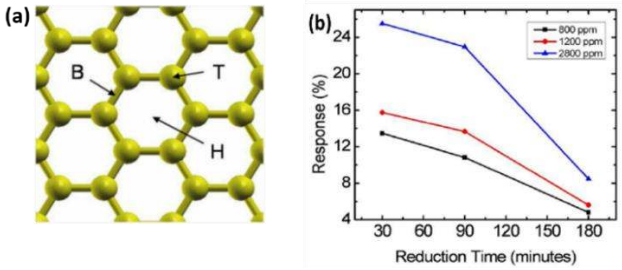

Fig. 8(a) Schematic showing three adsorption sites available in pristine graphene (reproduced from [77]), and (b) Response of RGO produced after chemically reducing GO for three different time durations towards three different concentrations of NH_3 (Reprinted with the permission from [67] Copyright©2013 American Chemical Society)

5.2 Intrinsic GO/RGO based gas sensors

Soon after the discovery of graphene, GO/RGO based gas sensor research also started. Robinson *et al.* demonstrated molecular gas sensors fabricated using RGO. They first synthesized GO and then reduced it chemically using hydrazine to get RGO which was used for sensing acetone and dinitrotoluene (DNT). It was demonstrated that RGO owing to its inherent defects and impurities is ultrasensitive towards gases and can sense gas species of concentration as low as 0.5 ppb [78]. Similarly, Lu *et al.* reported NO_2 sensing by thermally derived RGO [79]. GO contains many functional groups (epoxy, carbonyl, hydroxyl etc.). It was subsequently realized that these functional groups play major role in gas sensing, that is why GO performed better compared to pristine graphene as gas sensor. In fact, gas sensing properties of RGO are highly dependent on the extent of reduction of GO as shown in Fig. 8 (b) [67]. The report revealed that not only the response but response and recovery times of sensors are significantly dependent upon the reduction time of GO. The functional groups also render the sensing layer electrically insulating. Reducing GO helps in regaining the conductivity of the sensing layer but it also decreases the number of functional groups in it. So, there has to be a trade-off between the conductivity and sensitivity of RGO that can be achieved by optimizing the GO reduction time. The gas sensing mechanism of RGO is the same as that of graphene. But since RGO consists of functional groups which act as added attachment sites for the gas molecules so, in addition to physisorption, phenomenon namely chemisorption also occurs in RGO sensors. This theory has been computationally proven in the literature [80]. Our group demonstrated the above concept experimentally [67]. Chemisorbed gas molecules require more energy and time to desorb. So, the recovery of RGO is slightly slower than that of graphene. The recovery time of graphene is order of hundreds of seconds whereas the recovery time of RGO is usually tens of minutes. In recent years, GO has also been explored as sensor for detecting humidity. As the resistance of GO is very high so, the current across the sensors were observed in absence and then in presence of humidity. GO, owing to the presence of abundant functional groups, helps in high proton conductivity through attached humidity layers on its surface [63].

5.3 TMDs based gas sensors:

2D, layered, and semiconducting TMDs are also offering promise as potential alternatives of near room temperature gas sensing materials. Of different TMDs like WS_2, MoS_2, SnS_2, TaS_2, and TiS_2; MoS_2 based gas sensors have been explored the most so far [81]. All the other TMDs are still in embryonic stage but are expected to demonstrate excellent sensing results in the near future because of their outstanding properties. Ou *et al.* reported excellent NO_2 sensing by SnS_2 flakes. The sensors exhibited rapid response and recovery as the sensing was occurring due to physisorption of the gas molecules but the sensors were operated at high temperature (160°C) [82]. TMDs synthesized by liquid/chemical exfoliation have defects at their edges and also have sulphur (s)-vacancies. These sites offer to be the adsorption sites for the gas molecules. The adsorbing gas species donates/accepts electrons from the sensing layer which changes the conductance of the latter and hence the presence of gases are detected by these semiconducting 2D materials [53]. Table 1 enlists some of the recent works on intrinsic 2D layered materials-based gas sensing.

6. Drawbacks of pure 2D layered nanomaterials-based gas sensors

In general, 2D materials-based sensors suffer from a few fundamental limitations – first being their poor responses to gases. Although it has been shown in different reports that 2D materials are capable of sensing individual molecules but mostly such works were carried out under stringent lab conditions which differ a lot from the real-life circumstances. For example, in many cases the sensors were tested in presence of dry nitrogen and also long term reliability and reproducibility were not shown [75]. In real-life scenario, the sensors deployed to detect air pollutants are exposed to the environment that includes presence of several other gases along with humidity. Another drawback of such pure sensors are their slow response and recovery times [83]. The response and recovery times of efficient sensors should be not more than a few seconds. But most of the articles on intrinsic graphene, RGO, MoS_2, WS_2 or SnS_2 based sensors report response times and recovery times in order of few minutes or sometimes tens of minutes [67, 84]. Also, these gas sensors have other limitations like unstable response [85] and drifting

of the sensor resistance from its baseline value [86]. The next section of this review presents ways to overcome these problems.

Table 1: Key sensing parameters of intrinsic 2D layered materials based sensors reported recently

Sensing Material	Target gas(es)	Sensing temperature	Conc. Range	Response	Response time/ recovery time	Ref.
Epitaxial Graphene	NO_2	RT	1–50 ppb	1–17%	10–3000 s/Not reported	[87]
Crumpled RGO	NO_2	RT	1–10 ppm	R_{air}/R_{NO2} =1.5–3.8	8/53 mins	[83]
GO	NH_3	RT	2–100 ppm	6–30%	30 s/100 s	[88]
Multistep reduced GO	SO_2	27°C	5–50 ppm	5.93–47.44%	122/145 s	[89]
CVD MoS_2	NH_3, NO_2	RT	NH_3 (5–50 ppm) NO_2 (1.5–50 ppm)	NH_3<5% (50 ppm) NO_2 ~10 to 120%	Not reported	[13]
MoS_2 (UV activated)	NO_2	RT	5–100 ppm	~15–30%	29 s/Not reported	[84]
MoS_2	NH_3	75–150°C	50–480 ppm	10–70%(125°C)	650–700 s/750–1400 s	[53]
Chemically Exfoliated SnS_2	NH_3	RT	50–800 ppm	2.04–6.5 times	Not reported	[90]
WS_2	NH_3	RT	5–60 ppm	4.5–12 times	252 s/648 s (for 10 ppm)	[91]

7. Remedies to the drawbacks: Functionalization of 2D nanomaterials

One of the ways to overcome the limitations of nanomaterials described in section 6 is by functionalizing the nanomaterials using different species. This can be done in multiple ways as has been discussed below:

7.1 Functionalization with metal oxides

Metal oxides are well-known in gas sensing domain because of their ultra-sensitivity but have their own limitations as have been discussed already. 2D nanomaterials are capable of sensing gases at or near room temperature but they suffer from poor sensitivity. So, an obvious choice to confront the demerits of both these materials and to get the advantages offered by both the materials, is to form composites using these two sensing materials and to test its sensing capabilities. A good number of researches have been done in this direction and the concept has been experimentally proven. Our group demonstrated better sensing performance at room temperature by RGO–SnO_2 composite film for NH_3 as compared to the performance of the individual materials, as shown in Fig. 9 [92]. This is because metal oxides like SnO_2 are *n*-type semiconductors and RGO is a *p*-type material. So, when the composite is synthesized using RGO and SnO_2, it leads to the formation of hetero *p-n* junction. This junction offers an additional adsorption region for the gases that is not available with the individual materials; thus, a better sensing performance at room temperature is observed in the composites. Also, it is believed that RGO layer increases overall conductivity and hence measurable resistance is achieved even at room temperature. Various metal oxides including SnO_2 [93], ZnO [94], WO_3 [95], TiO_2 [96], CuO [97] etc. have been used to functionalize graphene/RGO and their sensing performance for different gases has been tested. Srivastava *et al.* presented a detailed investigation on RGO–WO_3 based NO_2 sensors. Although the developed sensors demonstrated optimum sensing performance at 250°C along with fast recovery and response times but the sensors exhibited decent sensing performance even at room temperature [98]. Recently, Zhang *et al.* reported CuO nanoflower/RGO based CO sensors. The composites were efficient enough to sense 0.25 ppm of CO at room temperature [99]. Lot of research on employing graphene/metal oxide composite for gas sensing is still going on. Few of such recently reported works are enlisted in Table 2.

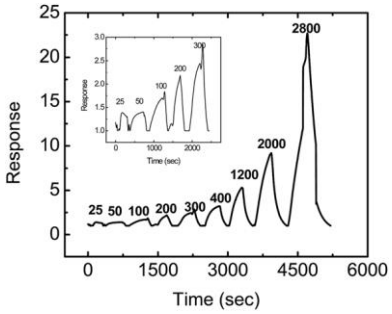

Fig. 9 Response of RGO-SnO2 sensing layer to different concentrations of ammonia (reproduced from [92] with permission of Royal Society of Chemistry)

Just like graphene and RGO, functionalization of TMDs using metal oxides have also been done and their performances as gas sensors have been investigated. Zhang *et al.* synthesized MoS_2-SnO_2 composite layer for ultrasensitive humidity sensing [100]. Similarly, our group reported enhanced gas sensing properties by WS_2–WO_3 composite layer [101].

7.2 Functionalization with metal nanoparticles

Along with other sensing materials, including metal oxides and 2D materials, sensing capabilities of some of the metals have also been explored over the years. Two of such metals are palladium (Pd) and platinum (Pt) because hydrogen molecules are dissociated on the

surface of both these noble metals thereby resulting in a shift in their Fermi levels, which changes their conductivities [102]. One of the demerits of metal nanoparticle based gas sensors is the cost. Usually, the sensors are fabricated by sputtering the metals over the substrates which increases the cost of the sensor because the sputter targets of these metals are very expensive. The cheaper solution is by reducing salts of metal (e.g. for Pt: $H_2PtCl_6.xH_2O$, for Au: $HAuCl_4.xH_2O$ can be used) in-situ while synthesizing the layered material [103, 104]. Such metal salts are comparatively less expensive than the highly pure sputter targets. In metal functionalized sensing layer, the gas molecules get three different types of adsorbing sites – functional groups or defects present in layered materials, surface of metal nanoparticles and the heterojunction formed between 2D materials and metals. Thus, improved gas sensing is exhibited at or near room temperature by these composites [105]. Conventionally, composites of 2D materials with Pt and Pd were explored the most because of the known reactivity of these metals with H_2. One such work has been reported by our group recently in which detailed investigation of the H_2 sensing performance of RGO-Pt based composite was carried out [103]. More recently research on functionalizing 2D layered materials with other metals like silver [106], iron [107], and nickel [108] are reported and their sensitivity towards different gases are being tested. For example, Huang *et al.* functionalized sulfonated RGO using silver nanoparticles and employed the composite for NO_2 sensing at room temperature. The Ag-S-RGO based sensors exhibited rapid response and recovery (each being few tens of seconds) [106]. The composites of TMDs and metal nanoparticles have also been synthesized and tested for different gases. For example, Baek *et al.* reported Pd functionalized MoS_2 based H_2 sensors(35.3% for 1% H_2 [105]), Burman et al reported Pt functionalized MoS_2 based humidity sensor (4000 times at 85% RH) [109]. A list of recent works based on metal functionalized 2D layered materials based gas sensors is given in Table 2.

Table 2: Key sensing parameters of functionalized 2D layered materials-based sensors reported recently

Sensing Material	**Target gas(es)**	**Sensing temperature**	**Conc. Range (ppm)**	**Response**	**Response time/ recovery time**	**Ref.**
RGO–MWCNTs–WO_3	NO_2	RT	1–5	9–17%	7/15 mins	[110]
ZnO–RGO	NO_2	RT	1–10	119–400%	75/132 s	[111]
RGO–CNT–TiO_2	Toluene	RT	50–500	42.9% (500ppm)	9/11 s (500ppm)	[112]
RGO– SnO_2	NH_3	RT	100–2000	R_{air}/R_{NH3} =1.014–11.79	8/13 s	[113]
RGO–TiO_2	NH_3	RT–100°C	1–50	1–4.9% (RT)	Not reported	[114]
CuO–RGO	CO	RT	0.25–1000	1.06–6.61%	70–76 s/147–232 s	[99]
RGO–ZnO	Chloroform, water, ethanol, acetone, formaldehyde	RT	Chloroform (20–80) Water (50–250) Ethanol (20–100) Acetone (20–100) Formaldehyde (25)	1.8–3.8% 2–6.1% 1.25–3.75% 1.25–3.75% 1.25%	<10 s	[115]
MoS_2–Co_3O_4	NH_3	RT	0.1–5	10–60%	105/136 s	[116]
Graphene–Ag	NH_3	RT	1000–12500	26–77.8%	120/72 s	[117]
Li–GO B–GO	Humidity	RT	11–97% RH	Li–GO (17.13–3038.16%) B–GO (6.95–631.1%	4/25 s 40/50 s	[118]
Graphene–Pd	H_2	RT	10000	30%	40/490 s	2017 SNB
Graphene–Polythiopene	NO_2	RT	1–10	~2.5–22.36%	Not reported	[119]
S-RGOH	NO_2	RT	0.2–2	6.1–22.5%	11/12 s	[120]
Graphene–Polyelectrolyte	Humidity	RT	10–90% RH	300–1000%	21 s/78 s	[121]
GO–ZnO–PANI	NH_3	RT	100	38.31%	30 s/Not reported	[122]
RGO–MoS_2	Formaldehyde	RT	2.5–15 ppm	1.5–6.5%	Not reported	[123]
Pd–TiO_2–MoS_2	Toluene	RT	100 ppb–100 ppm			[124]
MoS_2–GO	Humidity	RT	35–85%RH	3–1600 times	90 s/110 s (for 71.8%RH)	[125]
WS_2 – WSe_2	Humidity	RT	40–80%RH	15.4–57 times	45 s/65 s (for 60%RH)	[126]
SnS_2–SnO_2	NO_2	80°C	1–8 ppm	1.5–5.3 times	159 s/297 s (for 8 ppm)	[127]

7.3 Functionalization with Polymers

Another way to enhance the sensitivity of the 2D layered materials is to functionalize them using polymers. Polymers are organic molecules which consist of inherent functional groups and defect sites. So, in addition to the defect sites and functional groups present in the layered materials these added sites facilitate adsorption of more gas molecules on the sensor surface and thus perform better as gas sensors [128]. Development of polymer functionalized graphene/RGO based sensors has been well reported. Though most of the developed sensors are capacitive in nature [129], some research of polymer functionalized RGO based resistive sensors have also been reported. For example, Wu *et al.* in 2013 demonstrated enhanced NH_3 sensing by graphene/polyaniline nanocomposites at room temperature, which was attributed to the porous structure of the sensing material. They tested the sensors for a wide range of concentrations of NH_3 (1–6400 ppm) [130]. Lin *et al.* reported graphene/polypyrrole based humidity sensors which exhibited response times and recovery times in order of tens of seconds [131]. Humidity causes a lot of interference in sensors' performance. The authors of this review article found that if RGO is functionalized using Rose Bengal, which is an organic dye that finds wide utility in molecular electronics, then the sensitivity of the hybrid material enhances for NH_3 and that towards humidity reduces significantly [132]. Although the lifetime of polymers is often an issue as well as baseline drift, research into polymer functionalized layered materials is still being pursued because the composites can be synthesized using cost effective chemical routes and these composites have enormous potential as gas sensors. In 2016, Xie *et al.* reported MoS_2-P_3HT based ammonia sensors that were highly sensitive to low concentrations of ammonia. They tested the sensors for 4 – 20 ppm of ammonia and reported short recovery time [133]. Table 2 includes some of the recently reported works on polymer functionalized 2D layered materials-based gas sensors.

7.4 Mixed 2D layered materials-based sensors

Two different 2D materials having different sensing capabilities are expected to show improved performance. This is because the individual materials have their own defects and functional groups and when they are mixed, the advantages of the sensing capabilities of each material could be utilized. Also, the composites that have been so far been investigated are made of graphene and TMDs. Graphene/RGO is inherently *p*-type and TMDs are *n*-type materials. So, when graphene/RGO–TMDs are mixed, it results into formation of *p*-*n* junction. This heterojunction junction behaves as an additional site of attachment for the gas species thereby demonstrating a synergistic effect towards sensing applications [125]. In 2015, Cho *et al.* developed graphene/MoS_2 composite based sensors and demonstrated their NH_3 and NO_2 sensing capabilities at different operating temperatures including at room temperature. The sensors exhibited good responses towards the gases and could efficiently sense NH_3 and NO_2 down to 1.2 ppm concentration [134]. Our group also demonstrated enhanced humidity sensing by RGO/MoS_2 and WS_2/GO composite layers. The sensors exhibited better sensing compared to the individual materials. Also, the response times and recovery times of the hybrid materials were found to be very fast [66, 125]. The sensing performance of the WS_2/GO hybrid layer towards humidity is shown in Fig. 10 (a). The enhanced performance of the mixed 2D sensing layer is attributed to two phenomena occurring simultaneously in the sensing layer – (1) electron conduction through the sensing layer because of adsorption of water molecules and (2) proton conduction in which H^+ hops through the continuous water layer [125]. The humidity sensing mechanism by the mixed 2D material is shown in Fig 10 (b). Although not much research has been reported with the mixed 2D materials based resistive gas sensing, but this method has a lot of potential and more researches are expected to be carried out in the recent future.

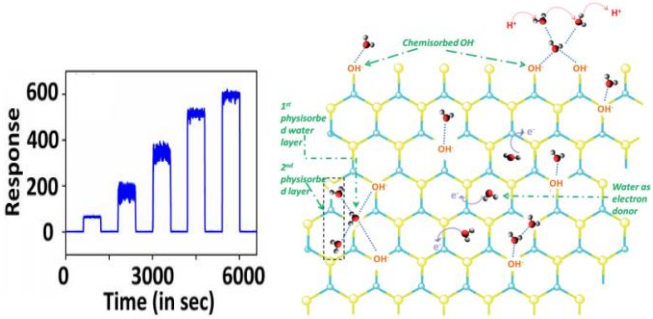

Fig. 10 (a) Response of WS_2/GO (1:3) hybrid layer for humidity [66] (b) Schematic representing proton conductivity as humidity sensing phenomenon in hybrid 2D material (reproduced from [125] with permission of Royal Society of Chemistry)

8. Pattern Recognition

One of the important parameters of sensors is selectivity. One way to improve this is by functionalizing the pristine 2D materials, but it is very difficult to achieve high selectivity in individual resistive sensors. This prompted the necessity to explore alternate ways of improving the selectivity of gas sensors. One such way is fabricating array of sensors, receiving signals from all of them simultaneously and then developing pattern recognition algorithms based on the sensors' response to accurately determine the gases present in the proximity and their respective concentrations [135]. This e-nose based approach has been widely reported [136]. This approach has now been widely reported and adopted for environmental monitoring (see 9.1).

9. Applications

There are enormous applications of gas sensors that range from environmental monitoring to process engineering and even the biomedical realm. Our review focusses on discussing the role of gas sensors for air quality monitoring which is required both at outdoors and indoors.

9.1 Environmental monitoring

Rapid industrialization, increased usage of automobiles and other human activities have escalated the emission of toxic gases like CO, NO_x, NH_3, ozone, SO_x and other greenhouse gases to hazardous levels. These gases have adverse effects on our health and wellbeing as mentioned in the introduction. Gas sensors can play a vital role in detection of these gases and in continuous monitoring. As achieving absolute selectivity in resistive gas sensors is not possible so, recent researches are more focused on development of array of gas sensors which are capable of recognizing multiple gases [128]. This led to

the evolution of new and advanced monitoring technologies like e-nose and e-tongues where e-nose is essentially an array of gas sensors employed to detect different obnoxious gases accurately and efficiently [137]. The major challenges in developing efficient air quality monitoring system for outdoor spaces are presence of humidity and wide range of deviation in the ambient temperature. These factors affect the sensor performance significantly. Research have been reported showing that 2D materials perform well at different temperatures and in presence of humidity [114]. Although there still remain rooms for improvement yet 2D layered materials like graphene, RGO and TMDs can be considered as the potential candidates to be employed for the development of efficient low power environmental monitoring systems.

9.2 Indoor air quality monitoring

Indoor air quality (IAQ) monitoring has become necessary today because most of us are required to sit inside an office and work for long durations (sometimes > 8 hours per day). In such circumstances if the indoor air quality is poor, then in many cases it leads to sick building syndrome (SBS) and might cause health hazards like headaches, dizziness and psychological disorders [138, 139]. IAQ deteriorates due to presence of gases like CO_2, VOCs like benzene, formaldehyde etc., microbial contaminants and particulate matters. The sources of these species are over-crowded confined spaces, inadequate ventilation, materials like permanent markers, cleaning supplies, glues, adhesives etc., and sometimes the building materials and furnishings. 2D layered nanomaterials-based array of sensors can be used to design alarm systems which would continuously monitor the quality of air inside the building and give signals when concentration of toxic gases exceeds safe limits.

10. Approach to reduce air pollution:

To detect air pollution efficiently, it is utmost important to develop low power low cost sensors (as is described above) that can be deployed in numerous sensor nodes. However, we feel the following points also need to be strictly adopted to give a chance for our future generation and perhaps a *right to breath*:

- As discussed above burning fuelwood for cooking contributes significantly to air pollution so, evolving with clean household fuel and making to accessible to common people would help improving the situation.
- Pollutants from automobiles can be controlled by enforcing tighter law and also by modernizing road infrastructure to reduce traffic congestion.
- Exploring renewable sources of energy and discovering the technologies to deploy the same efficiently would bring down the power plant emissions significantly.
- Precision agriculture which includes early detection of disease infestation and then precise and limited spraying of pesticides would help in reducing the pollution caused by agro-chemicals.
- Campaigns similar to BreatheLife (led by WHO, UN Environmental and the Climate and Clean Air Coalition) can be initiated by governments to spread social awareness about the effect on health due to air pollution and climate change.
- Concepts like smart cities and smart buildings with efficient monitoring systems and waste management should be designed and executed. Special care should be taken to preserve the natural assets like forests, rivers etc. while design the cities.
- Along with mass awareness, there should be several sensor nodes in smart cities (instead of only few fixed monitoring stations), internet of sensors, whose data will be accessible to general people (may be through mobile app) so that individual can make their travel plan to avoid pollution.

Conclusions

Our review discussed the different synthesis techniques of new 2D layered materials for gas sensor applications. We have reviewed the research that has been carried out so far on gas sensors based on intrinsic 2D materials, and explained the limitations of such 2D gas sensors. The advantages of functionalization of carbon nanomaterials and TMDs are also presented. Different ways of functionalizing these sensing layers are described and the performances of different composite sensors reported so far have been presented. The different components of a typical sensor device have been discussed. The final section of this review discussed some of the possible ways to minimize air pollution. It is believed that more focused research in developing gas sensors based on these new 2D materials could lead to the development of much more efficient AQM systems, which can reduce the 7 million deaths per year associated with very poor air as well as improve well-being for less polluted spaces.

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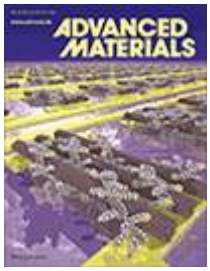

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