

Flexible Transparent Electronic Gas Sensors

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Flexible and transparent electronic gas sensors capable of real-time, sensitive, and selective analysis at room-temperature, have gained immense popularity in recent years for their potential to be integrated into various smart wearable electronics and display devices. Here, recent advances in flexible transparent sensors constructed from semiconducting oxides, carbon materials, conducting polymers, and their nanocomposites are presented. The sensing material selection, sensor device construction, and sensing mechanism of flexible transparent sensors are discussed in detail. The critical challenges and future development associated with flexible and transparent electronic gas sensors are presented. Smart wearable gas sensors are believed to have great potential in environmental monitoring and noninvasive health monitoring based on disease biomarkers in exhaled gas.

1. Introduction

Flexible, transparent, and conductive films (FTCFs) based electronics have attracted substantial research interest because of their potential application in integrated wearable electronics and display devices such as consumer electronics, homeland healthcare equipment, and environmental monitoring robotics.^[1] Recently, various FTCTF derived flexible

transparent electronic devices have been well developed, such as touch screens, electronic skins, environmental sensors, and wearable perspiration analysis platform.^[2] Among various applications, gas sensor is becoming an emerging area of critical importance due to the air pollution with the increased world-wide threat as well as exhaled gas with personalized diagnostic and physiological monitoring significance.^[3] Commercial electronics with FTCTFs as display components generally provide averaged Air Quality Index of specific regions downloaded from the internet. However, FTCTF based electronics still cannot provide timely gas pollution information in a specific location, which is much more important as the air quality varies greatly with time and place. In addition, wearable and portable sensor for monitoring exhaled gas is regarded as the economic and noninvasive technology for efficient disease detection as exhaled breath contains a variety of compounds at trace level concentrations serving as biomarkers for specific diseases.^[4] Therefore, it is significant to develop flexible transparent gas sensors (FTGS) capable of integrating into wearable electronics and display devices, which can provide real-time, sensitive, and selective gas analysis to timely and conveniently reflect environmental contaminant level or human physiologic status, as shown in **Figure 1**.

Advances in nanomaterial research have opened the door to flexible transparent conductive materials with gas sensing properties, mainly including semiconducting oxides, carbon based materials, and conducting polymers, all of which have

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demonstrated superior performance over laboratory-based bulky analytical techniques, such as chromatography and spectroscopy.^[5] Unfortunately, semiconducting oxides based traditional chemiresistive gas sensors generally rely on rigid substrates and require high operation temperatures of $>100^{\circ}\text{C}$, which pose certain drawbacks such as brittleness, high power consumption, and high cost due to its scarce supply.^[6] More importantly, the implementation of heating elements in sensor devices remains a major obstacle for the construction of miniature and integrated thin film sensors.^[7] These limitations have attracted an extensive effort to produce cheap, sensitive, and room temperature-operating FTGS. It has been shown that graphene and carbon nanotubes (CNTs) based detectors can operate at room temperature and readily form flexible films on plastic substrates.^[8] However, carbon derived sensors exhibited low detection sensitivity and sometimes required complicated recovery procedures such as current or UV light stimulus.^[9] Conducting polymers (CPs) are promising alternative candidates for developing flexible or wearable electronics, displays, sensors, and other devices, due to their outstanding electrical performance, tunable morphology, scalable manufacture, and low cost.^[10] However, CP based smart films still suffer from limited optical transparency, environmental instability, and require ceramic tubes or inter-finger probes to support traditional CP powders, rendering the fabrication of FTGS challenging. With the advantages and disadvantages of sensing materials in mind, rational design of nanocomposites with hierarchical structure may synergistically combine their advantages and avoid the weak points, thus resulting in the sensor devices with desired properties.

Based on the above illustration, an ideal smart sensor capable of being integrated in electronics should possess the following significant features: (i) flexibility and transparency; (ii) operation in ambient environment at room temperature; (iii) no external stimuli for fast response/recovery; (iv) low detection limit, high sensitivity, and reproducibility; (v) low cost and eco-friendly, etc. Despite ongoing achievements in this field, the former three features are still challenging and more efforts are needed to solve intractable scientific and technical problems. In this concept, we summarize the main advancements of FTGS based gas sensors, including carbon materials, metal oxide nanoparticles, CPs, and their composites based FTGS. Furthermore, we provide insights into the challenges and future development of FTGS to fulfill the future demands of wearable electronics and portable instruments with function of timely gas monitoring.

2. Flexible Gas Sensors

Nowadays, the growing demands for portable and wearable electronics have promoted increasing academic interest in flexible electronics.^[11] Conventional silicon based materials and organic semiconductors have been widely investigated for flexible electronics but their low stability and flexibility greatly limit the further practical application.^[12] Comparatively, low-dimension materials, such as CNTs, with excellent electrical and optical properties, higher carrier mobility and size-related intriguing physical properties, are becoming



and storage, stimuli-responsive biointerface, bioelectrocatalysis, supramolecular assembly, and nanoelectronics.

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promising materials in the construction of flexible electronics in terms of their room-temperature operability, rapid signal response, ease of use, and selectivity.^[13]

A typical CNT based flexible sensor was reported by Srikanth Ammu and co-workers, which was made from single walled carbon nanotube thin films on cellulose (paper and cloth) via inkjet-printed method (**Figure 2**).^[9c] Resistance changes were observed in relation to Cl_2 and NO_2 vapor with concentration of 5 to 100 ppm at room temperature due to the weak charge-transfer between CNTs and gases.^[9c] It was demonstrated that flexible substrate selection was an important factor for sensor recovery performance.^[9c] Porous cellulosic substrates facilitated the gas recovery in ambient environment at room temperature, while flexible polyethylene terephthalate (PET) substrates required photoirradiation treatment for recovery.^[9c] Despite the impressive flexibility of CNT based sensors, the signal response speed is too sluggish for practical real time application due to the weak adhesion of gas molecules on sensing materials. Motivated by this, Huan Liu and co-workers reported PbS colloidal quantum dots (CQD) on a flexible paper substrate via the layer-by-layer spin-coating deposition technique.^[5b] The porous thin CQD film of 80 nm guaranteed the rapid diffusion of gas, which enabled the rapid response at room temperature, high sensitivity, good reversibility, and outstanding mechanical bending ability.^[5b] In brief, rational design of one dimensional materials or patterned nanoparticles with excellent sensing properties and good adhesion to flexible substrates is a feasible strategy for flexible sensor construction.

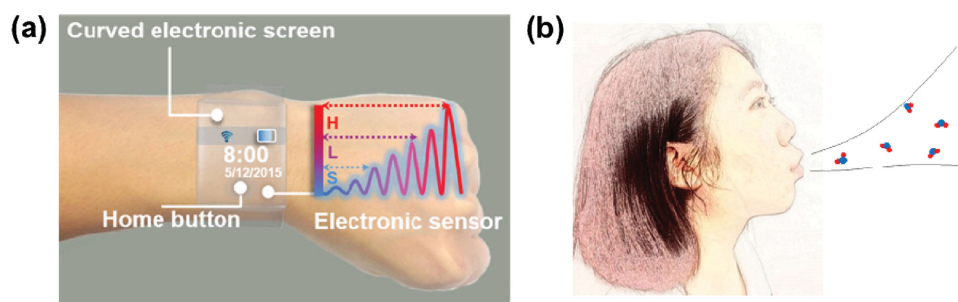


Figure 1. a) Scheme of a flexible transparent electronic sensor integrated into wearable devices detecting environmental gas, or b) exhaled gas, indicating the air pollution level or human physiologic status, respectively (S: safe, L: low toxicity, H: high toxicity).

3. Transparent Gas Sensors

Numerous electronic devices including mobile phones, smart watches, kindles, liquid crystal displays, have common digital screens, which use a material with high transparency to visible light as an essential element in the device.^[7a] The key step to enable the integration of sensors into electronics is to develop optically transparent sensors. Traditional semiconductor metal oxides based chemiresistive chemical sensors generally require the integration of heating elements to insure the rapid and reversible adsorption and desorption of gas on the surface of the semiconducting materials.^[6a,7b] The presence of heating elements greatly hinder the construction of transparent sensors.

Aimed at solving the above challenges, transparent sensors based on self-activated metal oxide thin film were reported, which avoided the use of a heating element in the gas sensor.^[14] The self-activated sensor was based on electronic self-heating WO_3 thin films with nanocolumnar structure.^[14] Nanocolumnar WO_3 films were fabricated via firstly patterning indium-tin oxide (ITO) interdigitated electrodes (IDEs) on glass substrates and then deposition of porous WO_3 thin films onto the IDEs (**Figure 3a**).^[14] Self-heating in the WO_3 film originated from the unusual geometry of the porous nanocolumnar structure (**Figure 3b**), where electron flow was constricted, leading to increased joule heating.^[14] On the other hand, ambient air in the pores provided an excellent thermal barrier, which suppressed heat dissipation laterally to the IDEs, and the small contact area between

nanocolumns and glass substrate minimized heat dissipation to the substrate.^[14] The temperature of films rose to 139 °C at the applied bias voltage of 5 V (**Figure 3c**), illustrating the high efficient electric self-heating property.^[14] The nanocolumnar sensor showed high transmittance of 90.2%, excellent NO_2 sensing performance (detection limits of parts per trillion levels), and low power consumption (less than 0.2 microwatts).^[14] This exhibited great advantages over the dense planar sensor which has negligibly small and sluggish response due to the latter's low sensing temperature and reduced active surface area (**Figure 3d and 3e**).^[14] Even though the self-activating transparent sensor avoided an extra heater element, the gas detection process was still conducted at high temperature with potential safety issues and the construction of nanocolumnar structure was rather complicated.

Alternatively, CPs, such as poly(3,4-ethylenedioxythiophene) (PEDOT) and polyaniline (PANI), have been widely studied in developing flexible or wearable electronics due to their tunable optical, electronic, and chemical properties.^[10,13a,15] In particular, PANI demonstrates several advantages such as facile synthesis, environmental stability, unique acid/base doping/dedoping, and oxidation/reduction property, and therefore has been proven as a promising candidate in sensing applications.^[16] However, most of the gas sensors are based on solid powders materials and the reported PANI films still have drawbacks such as unsatisfactory gas sensing performance, limited optical transmittance, and high electrical sheet resistance. Our group reported a transparent gas sensor, which is totally conducted at room temperature based

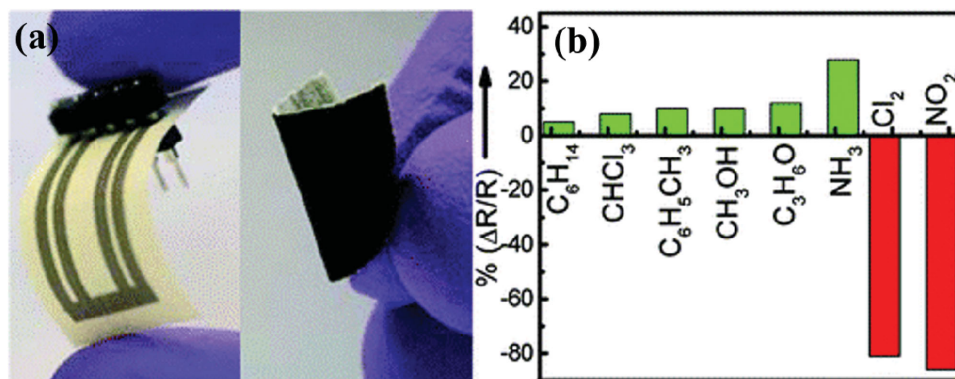


Figure 2. a) Flexible chemiresistors based on CNT films on cellulose. b) Selectivity plot for an inkjet-printed CNTs/PET film toward saturated organic vapors, NH_3 (100 ppm), NO_2 (100 ppm), and Cl_2 (100 ppm). Reproduced with permission.^[9c] Copyright 2012, American Chemical Society.

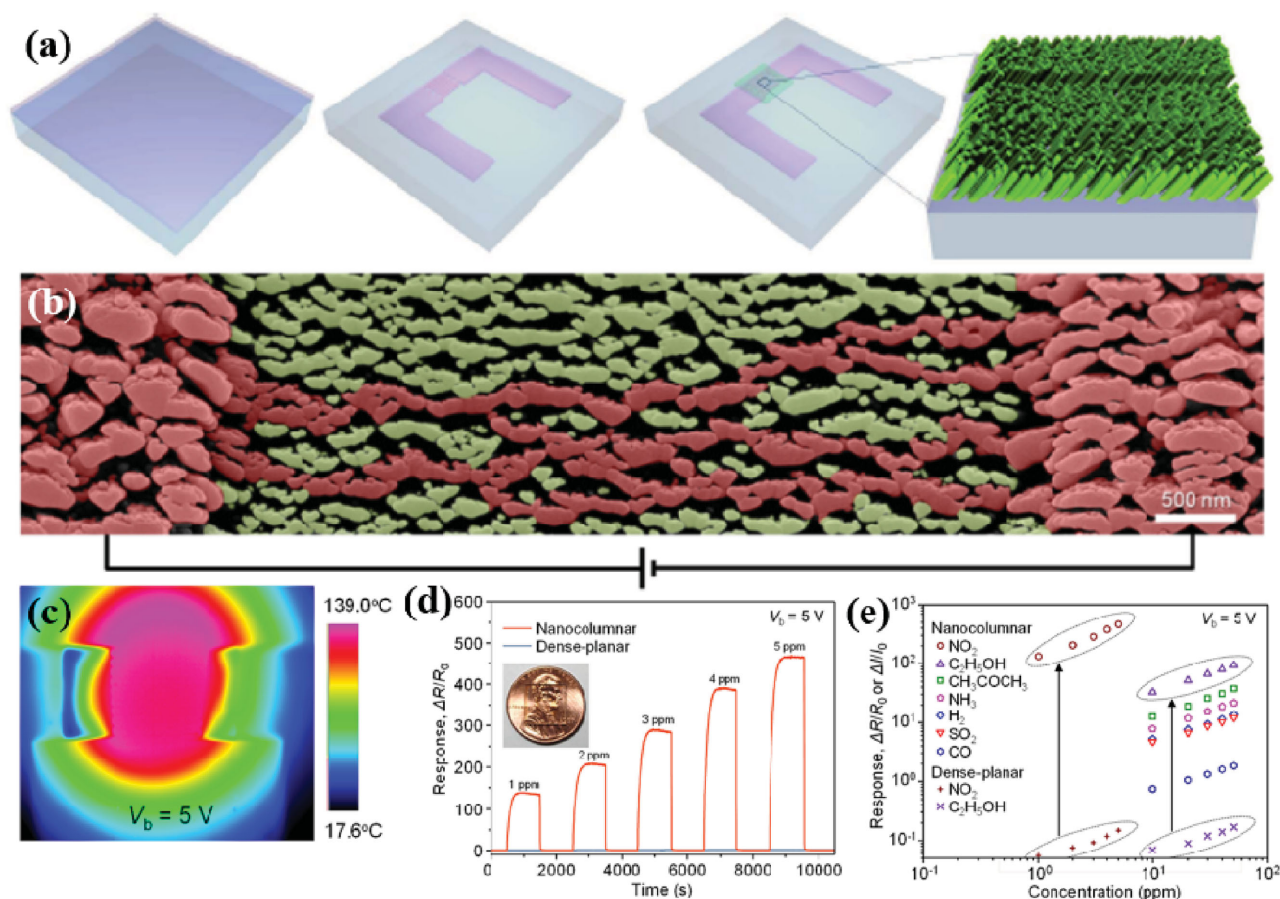


Figure 3. a) Schematic fabrication of nanocolumnar oxide films based transparent sensors. b) 40°-tilted SEM image of nanocolumnar WO₃ film. Parts highlighted in reddish color indicate localized and meander current pathways with narrow necks. c) Thermographic image showing temperature variation in the WO₃ thin film sensors with bias voltages of 5 V. d) Sensing transients of the dense-planar and nanocolumnar WO₃ thin film sensors to 1–5 ppm NO₂ at a bias voltage of 5 V, the inset: photograph of a sensor on a coin. e) Response of the nanocolumnar WO₃ thin film sensors for various target gases. Reproduced with permission.^[14] Copyright 2012, Nature Publishing Group.

on hierarchically one dimensional (1D) nanostructured PANI networks on a PET substrate.^[17] Exploiting Ag nanowires (AgNWs) on PET as the sacrificial template, the in situ oxidation polymerization of aniline produced transparent PANI networks films (Figure 4a).^[17] The sensing mechanism is as follows: the deprotonation of acid-doped PANI by NH₃ caused a dramatic decrease in conductance by changing PANI's emeraldine salt form into its emeraldine base form (Figure 4b).^[17] It is noted that the hierarchically nanostructured PANI-containing film with optimal polymerization time and good transparency had higher gas sensitivity and selectivity than the PANI particles based film ascribed to the quasi-1D nanostructure in the PANI network structure with large specific surface area and the specific acid-base reaction (Figure 4c and 4d).^[17] Owing to their high transmittance, the visible sensors could be embedded in next-generation smart displays equipped with transparent electronics for aerospace, military, automobile, and biomedical applications.

4. Flexible Transparent Gas Sensors

The development of flexible sensors to substitute the rigid ones in smart devices should solve the application bottleneck

of smart devices, where flexibility, carrier mobility, size reduction, or production constraints limited their serviceability. The high transparency of sensors in electronic devices allows the displays of touch screens, flat panel displays, image recorders, optical communication devices, and solar cells to function normally. However, rigid transparent sensors are generally adversely influenced by mechanical deformation due to the readily produced rifts in sensor platform, which greatly limits the application of transparent sensors. Therefore, both mechanical flexibility and transparency are essential properties to be considered in order to integrate sensors in portable and wearable electronics, which have already driven substantial research efforts in this area.

Most recently, Zheng and co-workers prepared a flexible and transparent ethanol gas sensor based on ZnO nanoparticles using ITO coat PET as the substrate which was capable of working at room-temperature.^[18] Despite the transmittance of 62%, satisfactory flexibility with curvature range of 90°, and rapid response without heating elements, the ZnO nanoparticle sensor required UV irradiation, which complicated the sensing process and was undesirable for wearable devices.^[18] Exploiting the excellent electrical and thermal conductivity of graphene, a graphene based flexible and transparent sensor was fabricated, including a graphene sensing channel

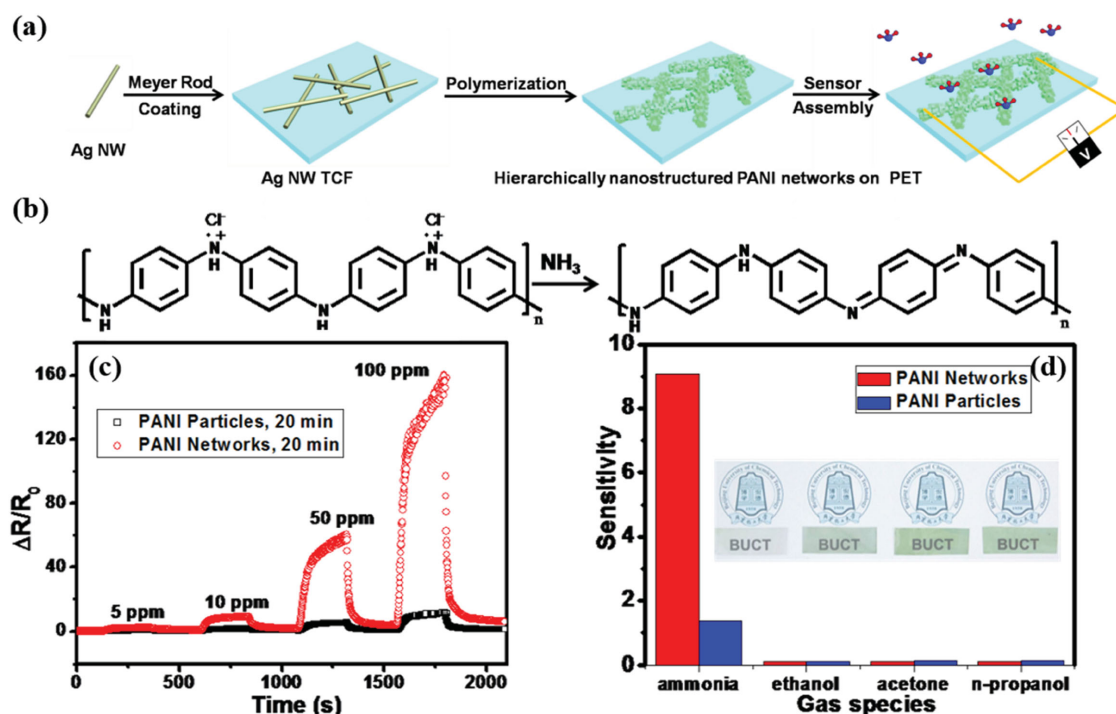


Figure 4. a) Scheme of fabrication of PANI networks based transparent conducting film on PET substrate. b) Sensing mechanism of acid-doped PANI toward NH_3 . c) The gas sensing of the PANI network-containing transparent film (red) and the particle-like PANI (black) upon exposure to NH_3 with concentrations varied from 5 to 100 ppm at room temperature. d) Gas sensing selectivity test of the PANI network-containing transparent film and the particle-like PANI film to various volatile organic gases with concentration of 10 ppm. The inset: photograph of PANI network films on PET substrate with the immersion time at 5, 10, 20, and 30 min from left to right.^[17] Copyright 2015, Wiley-VCH.

and a graphene heater.^[19] Using polyethersulfone (PES) as the transparent and flexible supporting layer, single-layer graphene (SLG) and bilayer graphene (BLG) were fabricated at the center and sides of the PES substrate, which served as a sensor channel and heater, respectively (Figure 5a and 5b).^[19a] The center of the channel exhibited a temperature deviation between 100 to 165 °C under the supplied heater temperature of 250 °C (Figure 5c), which was sufficient for the rapid detection and recovery step.^[19a] With increasing uniaxial tensile strain applied from 0 to 1.4% along the longitudinal (*y*-axis) directions, gas sensing and recovery cycles varied slightly with a $\Delta R/R_0$ deviation of less than 5% (Figure 5d and 5e), indicating the satisfactory flexibility.^[19a] This flexible and transparent all graphene sensor and heater device exhibited high sensitivity with a $\Delta R/R_0$ ratio of 10% towards 0.5 ppm NO_2 gas, high optical transmittance ($T_r > 90\%$), high bending strain resistance, and a rapid recovery time smaller than 20 s, which illustrated the possibility for an unconstrained size and flexible device integrated in smart windows and wearable nose-sensors.^[19a] Despite their promising advantages, there still needs improvements in flexibility. The bending direction was only confined to *y*-axis to ensure the good performance. Bending in the transverse (*x*-axis) direction may produce rifts in the juncture of graphene sensor and graphene heater, thus damaging the connections and eventually leading to a decrease in performance, even the breakdown of sensors. Furthermore, micropatterned graphene sensor with self-activated function was designed to enable stable contact between graphene and sensor electrodes.^[20] The bent sensor showed only $\approx 3\%$ performance degradation under bending

radius of 1 mm.^[20] Self-activation was based on Joule heating in a micropatterned graphene channel attributed to sensor geometry and applied bias voltage.^[20] Self-activated all-graphene sensors exhibited high transparency, excellent flexibility, and low power consumption, which are promising as components in hand-held devices such as mobile phones, smart watches, and laptops.

Even though the strategy of using a transparent graphene heater partly solved the problem of opacity due to the heater element in sensor, the sensor and recovery process was still conducted at temperature higher than 100 °C. Consequently, transparent and flexible sensors at room temperature were highly desirable for practical application. Our group constructed a flexible and transparent sensor based on nanostructured network of PANI-coated CNT on PET substrates.^[21] The polymerization of aniline in the CNT suspension produced PANI/CNT composites, which were concurrently deposited on a PET substrate and yielded PANI/CNT nanocomposite films in situ (Figure 6a).^[21] The assembled device using PANI/CNT film displayed a satisfactory transparency of 85.0% at 550 nm, flexibility (without significant performance degradation after 500 bending cycles), high selectivity, portability, and room temperature operating conditions (Figure 6b and c).^[21] It was illustrated that the flexibility, transparency, and good sensing performance were ascribed to the quasi-one-dimensional nanostructure CNT/PANI composites network, which synergistically combined the superior properties of both CNT and PANI, including the high surface area and reliable NH_3 sensing property.^[21] The research provided a low-cost, sensitive, and reliable sensor

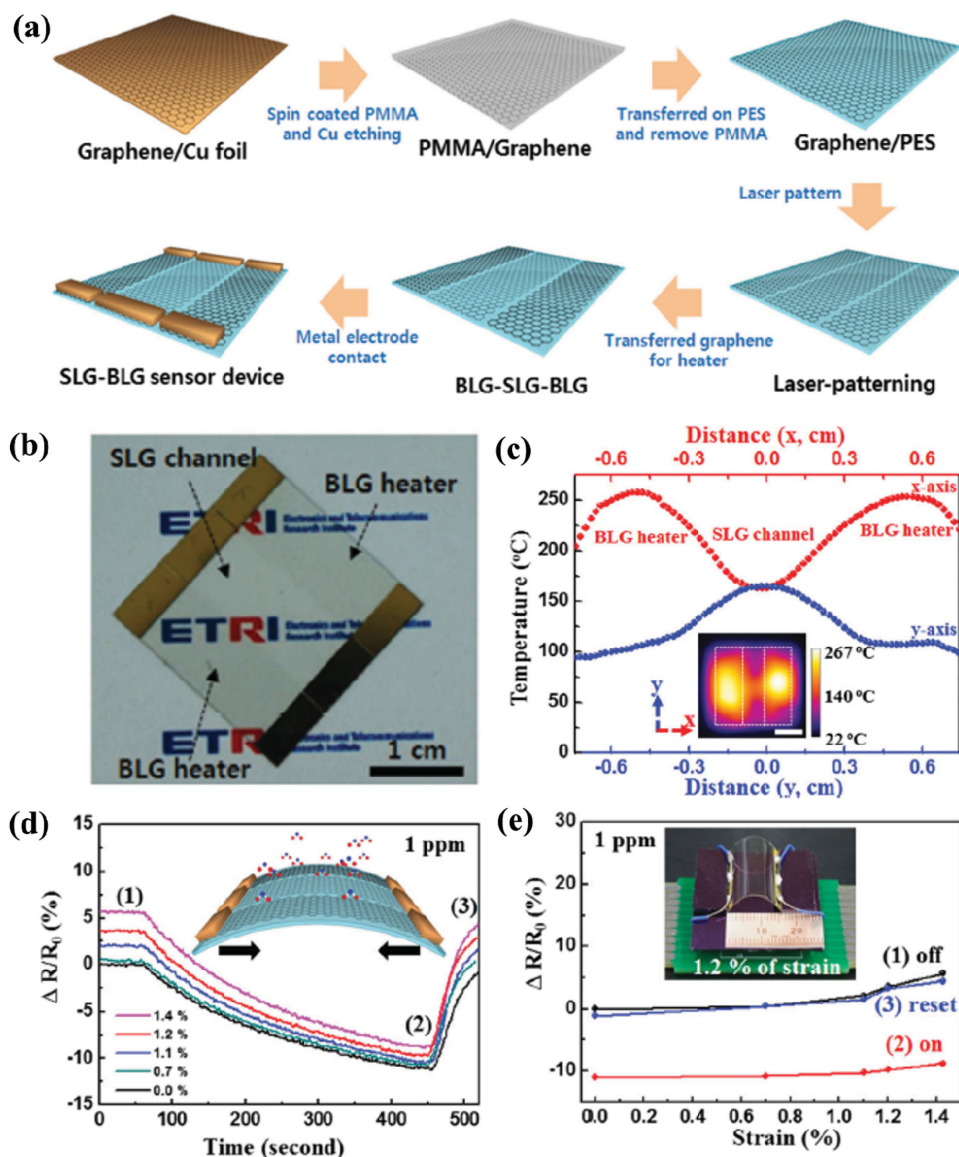


Figure 5. a) Scheme of fabrication for laterally positioned graphene sensor and heater on flexible transparent substrate. b) Optical microscope image of transparent flexible SLG sensor channel-BLG heater on PES substrate. c) Temperature distribution along transverse and longitudinal direction of sensor-heater device structured with applied 1.7 W of electric power. d) The $\Delta R/R_0$ of SLG channels as a function of time including recovery step under NO_2 (1 ppm) with 0 to 1.4% bending strain at y-axis direction. Inset: Schematic diagram of strained graphene sensor in NO_2 gas condition. e) The $\Delta R/R_0$ of sensor at initial, saturated, and fully recovered conditions denoted as 1) off, 2) on, and 3) reset at (d), respectively, under 1 ppm NO_2 gas as strain varied from 0 to 1.4%. Inset: Optical image of bended graphene gas sensor-heater junction under 1.2% bending strain. Reproduced with permission.^[19a] Copyright 2014, Wiley-VCH.

with the potential to be integrated into portable, wearable, and transparent electronic devices.

Furthermore, due to the commonly existence of mechanical deformation or accidental cutting in the practical applications, healable flexible and transparent sensors attracted great attention. Inspired by this, a healable transparent chemical gas sensor was designed by our group through casting transparent CNT networks on healable polyelectrolyte multilayer films (PEM) prepared via layer-by-layer assembly route.^[22] Due to the interdiffusion and the flowability of PEM substrate, the healable function was achieved via deionized water induced lateral movement of the PEM films, which brought the cut areas of the conductive CNT layer back into

contact.^[22] Healable and transparent sensors possess extra advantages including the decreased raw material consumption, reduced maintenance costs, enhanced lifetime, and robust functional reliability, which is anticipated to further broaden the application of flexible and transparent sensors.

5. Limitation, Challenges, and Opportunities of Flexible Transparent Sensors

To enable the high flexibility and transparency of sensors, the following issues should be simultaneously addressed: (i) the flexibility, transmittance, and permeability of substrates;

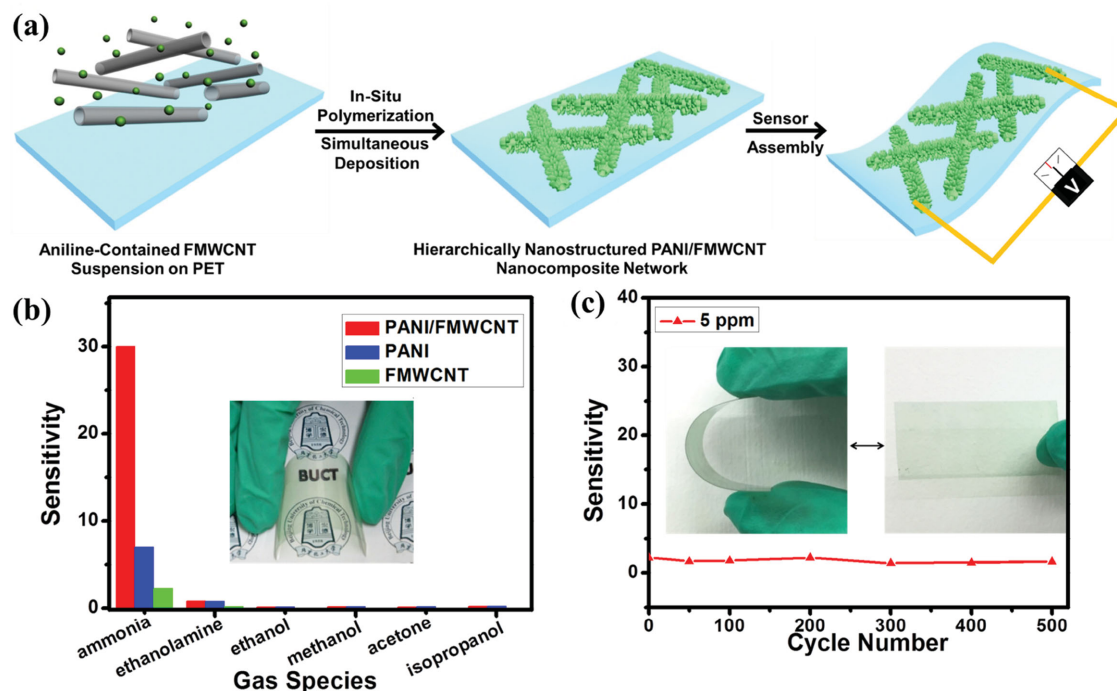


Figure 6. a) Scheme for the preparation of the hierarchically nanostructured PANI/CNT nanocomposite network film for flexible transparent chemical gas sensor. b) Gas sensing selectivity of PANI/CNT nanocomposite, the particle-like PANI aggregate and the CNT film based devices. Inset: Digital image of the flexible transparent PANI/CNT network based film. c) Chemical gas sensing performances of PANI/CNT nanocomposite network film in bending and extending states to 5 ppm NH_3 after different bending cycles.^[21] Copyright 2015, Wiley-VCH.

(ii) the flexibility, transmittance, microstructure, and adsorption property of sensing materials, (iii) the uniform distribution configuration of sensing materials on substrates. As mentioned above, using PET, polyimide (PI) or cellulose as substrates and patterned metal oxide NPs, 1D/2D carbon materials, quasi 1D polymers or hierarchical composites as sensing materials, FTGSs with impressive performance in the sensitivity and response/recovery time have been achieved. Chemiresistive sensing mechanism of FTGSs can be classified as (i) the interaction of the oxidizing or reducing analytes with surface atoms, inducing charge redistribution in the p- or n-type semiconductive materials, such as PbS CQD based NO_2 sensor,^[5b] PANI based NH_3 sensor,^[17,21] ZnO NPs based $\text{C}_2\text{H}_5\text{OH}$ sensor,^[18] etc. (ii) the permeated volatile organic compounds (VOCs) vapor-induced swelling of films, leading to a reduction in the conductive pathways, such as capped AuNPs for detecting ethanol, chloroform, hexane, and toluene.^[3b,23] For the detection of a certain gas, one mechanism plays a dominant role or the two types of mechanism synergistically contribute to the response, which is dependent on the interaction between analytes and sensing materials. Nevertheless, due to the similarity in functional groups and property of various gases, the selectivity and reliability in a complex environment are still major impediments to the realization of FTGS in real-world applications.

One challenge of FTGS is the limited selectivity and low selectivity towards CO and VOCs such as various hydrocarbons and solvents, which greatly hinders the widespread application of FTGS. Current metal oxide based CO, ethanol, trimethylamine and HCHO sensors show good performance

but need high temperature to activate the interaction between gases and adsorption sites on metal oxide.^[24] For solving this problem, nanoparticles were effectively capped with functional groups, monolayers, polymers, or biomolecules to selectively detect a specific analyte.^[23b,25] CNT/dodecanethiol-capped-AuNPs composites based flexible PI film was reported as high sensitivity to trichloroethylene and chloroform at room temperature due to the capped alkyl chains with a higher tendency to nonpolar VOCs.^[13j] For large and less reactive VOCs such as benzene, xylene, and toluene, the reported sensors at high temperature generally exhibit limited sensitivity with linear detection range of ppm level due to weak/lack of interaction of analytes.^[26] Recently, p-n heterojunctions based on polyaniline-SnO₂ composite materials showed room temperature sensing performance toward 10% of benzene/toluene gas due to the physisorption of benzene/toluene on PANI, which attracted electrons from p-doped PANI, perturbed electron density and thus reduced the conductivity.^[27] However, the sensor still failed to determine the identity and quantify the amount of each VOC in a complex mixture due to the structure and property similarity of benzene and toluene. Up to now, rare FTGS toward VOCs was achieved and thus FTGS with high sensitivity and selectivity towards VOCs was still in its infancy. When transferring current VOCs sensors into FTGSs, there is a dilemma that reducing film thickness increases transparency and flexibility but decreases sensor performance. Moreover, most of the VOCs sensors use inorganic nanoparticles powders, which are difficult to transfer into uniform thin, transparent and flexible films due to the

low transparency and limited interaction of inorganic nanoparticles towards flexible substrates. Therefore, rational design of target-analyte-specific materials with satisfactory flexibility, transparency, selectivity, and sensitivity is still a promising but challenging route to broaden the real application of FTGS.

The as-reported FTGSs have been demonstrated only under controlled laboratory conditions, while real applications comprise a range of chemicals and various environmental changes. Humidity effect is a typical adverse factor. H₂O adsorption-induced performance deterioration was generally observed for semiconducting metal oxides and polymer based gas sensors, which needed high temperature to alleviate the water poisoning effect.^[28] Recently, graphene based self-activated sensor exhibited humidity resistant performance and it was noted that self-activated property based on Joule heating played a role in reducing the humidity effect, which avoided the use of extra heater elements.^[20] However, the essence of strategies to resist humidity increased the temperature to enable the replace of the adsorbed H₂O with target molecules, which brought about power consumption and safety issues for real applications. Consequently, sensing materials with humidity resistance property are highly desirable but still challenging to construct a reliable and real-world applicable sensor.

6. Conclusions and Perspectives

Flexible and transparent electronic sensors have bright prospects for the application in smart electronics. Recent progress in the fabrication of FTGS based on semiconducting oxides, carbon materials, conducting polymers, and their nanocomposites is highlighted. The carbon material based flexible sensor has the drawback of limited sensitivity while the transparent sensor constructed from semiconducting oxides generally needs high operation temperature to enable the rapid gas adsorption and desorption process, both of which failed to construct flexible and transparent room-temperature sensors. Comparatively, hybrid nanocomposites synergistically combines the merits of individual elements, which are desirable choices, such as conducting polymer and CNT hierarchical nanocomposite network based electronic films with excellent flexibility, high transparency, and room temperature operation conditions. Even though, it is still challenging for the reported FTGS to provide the reliable identity and quantity information of target gas at room temperature in a complex environment, especially for large and less reactive VOCs.

To promote the further development and wider applications of FTGS, following directions should be highlighted. Persistent efforts should be devoted to searching for flexible and transparent platforms composed of sensing materials with high sensitivity, stability, and selectivity at room-temperature regardless of sensing condition variations. Based on reported sensors composed of flexible sensing materials, selecting flexible and transparent substrates, uniformly distributed sensing materials on substrates with desired patterns should be a possible way to construct various FTGSs. In addition, smart FTGS with multi-functionality, such as readily

healable or stretchable ability is the next goal of this field in order to minimize the possibility of mechanical fracture in practical application, which should greatly improve their reliability and lifetime, reduction in raw material consumption and maintenance costs. More importantly, with the presence of disease biomarkers in the human exhaled breath, the development of skin-conforming FTGS into wearable devices will potentially provide a painless and non-invasive method to timely monitor an individual's state of health via simple exhalation toward the wearable devices. The last but not the least, FTGS was generally confined to one type of gas species, it is interesting to integrate various FTGS into lab-on-chip devices, which thus can simultaneously provide various gas concentration information and an overall air quality evaluation system in real-world application. By solving the scientific and technical issues described above, we believe that flexible and transparent electronic sensors will promote a new generation of smart portable electronics and wearable devices.

Acknowledgements

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