

Structural Effects of Crumpled Graphene and Recent Developments in Comprehensive Sensor Applications: A Review

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Graphene is a 2D honeycomb lattice consisting of a single layer of carbon atoms. Graphene has become one of the most preferred materials for sensor development due to its exceptional electrical, mechanical, and thermal characteristics. Nonetheless, little consideration is given to the production and use of crumpled graphene. Specifically, the crumpled graphene structure is a good choice for enhancing sensors' sensitivity and structural deformability by reducing interfacial stress, avoiding electrical failure, and enhancing surface areas. This review article provides an overview of various synthesis processes using crumpled graphene and specifies a brief idea to control crumpled formation in graphene structure for sensing applications in recent years. Furthermore, it summarizes the problems encountered in previously published research articles during the fabrication and performance of sensors with a brief discussion of fundamental mechanics and topological aspects concerning crumpling patterns with sensing performance. It also highlights the current status of crumpling techniques and their effects on developing different sensors using existing crumpling methods, controlled crumpling designs, and sensing methodologies for future applications.

increasing due to its stability and impressive electrical and mechanical attributes: mobility of charge carriers ($250\,000\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$) at an ambient temperature,^[5] heat conductivity ($5000\text{ W m}^{-1}\text{ K}^{-1}$),^[6] and mechanical stiffness = 1 TPa .^[7] However, producing graphene in large numbers and various configurations using basic mechanical and chemical methods remains challenging.^[8]

Graphene was first discovered by mechanically peeling bulk graphite using Scotch tape, also known as the micromechanical cleavage technique.^[9] However, this mechanical exfoliation process is neither high throughput nor high yielding, making it unsuitable for most industrial applications.^[10] In contrast, chemical vapor deposition can produce excellent-quality graphene with a controllable quantity of layers and a large surface area. High-temperature metal catalysts facilitate the decomposition of precursor materials into carbon atoms, which combine to produce

graphene.^[11–19] However, due to high intersheet adhesion, the tendency of graphene to agglomerate (restacking) is a significant concern. Restacking nanosheets reduces their available surface area, thus helping in altering their properties and possible applications to transform 2D structures into 3D crumple structures.^[20,21] The curvature of a 3D graphene sheet caused by external confinement or pressure mainly concentrated on the ridges could significantly change the characteristics of the material.^[22] Numerous researchers have studied the generation of ripples, folds, wrinkles, or crumples on graphene sheets and the use of such corrugations in a controlled manner.^[23–31] Wrinkling is a relatively common physical phenomenon in thin sheets and membranes.^[32,33]

Due to its potential uses in sensors and capacitors, developing graphene crumple structures is one of the most important current topics in cutting-edge materials. At room temperature, fabricated nanosheet gas sensors exhibit an effective NO_2 response. Notably, the simplicity and cost-effectiveness of the lyophilization-induced process for creating 3D CG-based nanosheets have raised the possibility of mass-producing gas-sensing materials.^[34] Creating CG from regular graphene requires altering the shape and structure of graphene using a prestretched thin layer of silicone. The remarkable strain sensitivity of CG sheets indicates their great potential for applications in strain gauge devices.^[35] The polydimethylsiloxane (PDMS)-based CG strain

1. Introduction

Graphene, a revolutionary 2D carbon substance discovered a decade ago, has attracted considerable interest owing to its extraordinary capabilities stemming from its unusual shape.^[1–3] The value of the global graphite market is expected to be $\approx \$27$ billion by 2025 (Figure 1), an increase of more than \$9 billion from the 2018 estimated market value.^[4] Graphite applications are continuously

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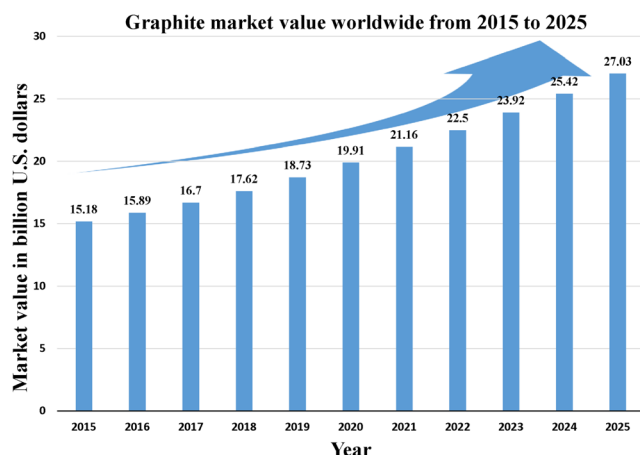


Figure 1. Worldwide, Statista estimates; Adroit Market Research; 2015 to 2018. Reproduced with permission.^[4] Copyright 2019, Statista.

sensor exhibits satisfactory biocompatibility. Despite hundreds of stretch–release cycles, such sensors could sense massive strain deformation and retain good performance.^[36] Song et al. developed a simple synthesis method for obtaining CG oxide (CGO) and demonstrated the exceptional electrochemical performance of the CGO electrode as an anode. They also discussed the

morphological modification of GO to CGO for enhanced electrochemical performance.^[37] The wrinkle/wrinkled sensing layer has shape-modulating capabilities,^[38] while soft substrates are deformed.^[39] Additionally, thin sheets (wrinkled structure) released interfacial tension, reduced electrical failure, and increased the surface area, leading to graphene-based energy storage device application.^[40,41] Integrated sensor devices based on surface crumpling/wrinkling may thus exhibit remarkable deformability and sensing capability.

This review paper focuses on the kinematics and sensing applications of crumpled forms in 2D graphene structures. **Figure 2** depicts several crumpled surface formations, designs, features, and applications. The fabrication mechanics with controlled crumpling fabrication methodologies for crumpled shapes are introduced along with associated structural applications (e.g., sensors with high flexibility and cutting-edge applications), along with the constraints, difficulties, and prospects of CG in sensors are examined.

2. Synthesis Method

2.1. Thermal Reduction Method

The material must be free of oxygen and reactive chemical species to get improved design and quality of crumpled structures

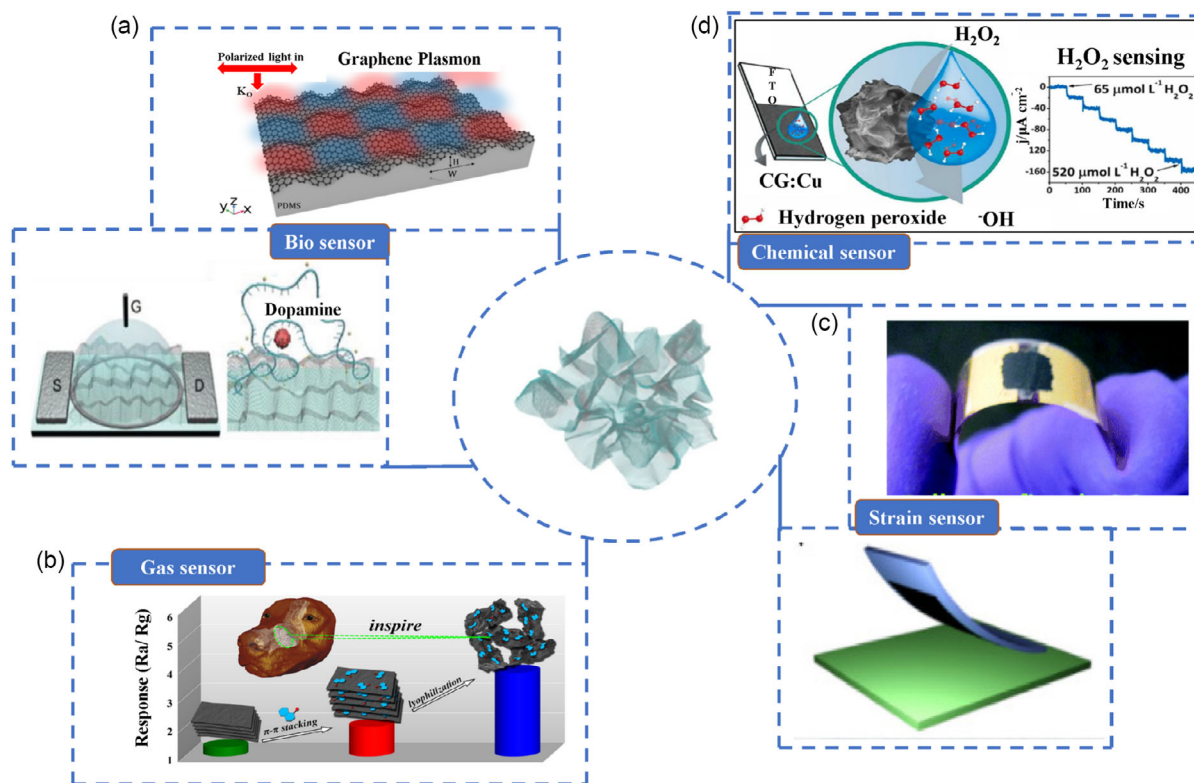


Figure 2. Applications of crumpled graphene. Reproduced with permission.^[21] Copyright 2011, American Physical Society. a) Biosensor. Reproduced with permission under the terms of the Creative Commons CC BY license.^[130] Copyright 2022, the Authors. Published by MDPI. Reproduced with permission.^[128] Copyright 2021, Wiley-VCH. b) Gas sensor. Reproduced with permission.^[34] Copyright 2017, American Chemical Society. c) Strain sensor. Reproduced with permission.^[161] Copyright 2017, Royal Society of Chemistry. Reproduced with permission.^[147] Copyright 2017, Elsevier. d) Chemical sensor. Reproduced with permission.^[154] Copyright 2022, Elsevier.

because their presence could lead to carbon loss caused by the production of gases or volatile compounds in graphene. In addition to eliminating graphene oxide (GO), rapid heating diminishes functionalized graphene films by dissolving oxygen-incorporated groups at high temperatures. The thermal degradation of GO is a conventional heating process that involves slowly heating a solid mass of GO or a mixture of GO and liquids in an inert environment. Because of its effects, the thermal reduction of GO has become the preferred method of graphene production. However, this method has several disadvantages because it can only manufacture tiny, wrinkled graphene sheets, primarily due to the breakdown of oxygen-containing sets and the elimination of particular carbon atoms from the graphene backbone.^[42] Researchers prefer thermal treatment over chemical treatment since chemical treatment necessitates using hazardous chemicals and is time intensive.

The thermal degradation process requires a simple setup and easy steps.^[43,44] It is the preferred method because it is easy to perform and produces better products. However, the kinematics of the heat reduction process involved in creating GO must be explained. Because the thermal degradation of GO might generate an explosion, caution should be taken while heating samples. In the article, the author elaborated on the catastrophic behavior that is more likely to occur, and the initial temperature for the GO degradation releases heat upon immersion in KOH.

Further studies are required to comprehend the kinetic parameters and reaction mechanisms of the reduction process to adequately distinguish the potential dangers associated with massive GO processing and allow for its protected development and commercialization, as the research available to date is insufficient to draw any firm conclusions.^[45] Farah et al. obtained thermally reduced GO (TRGO) by moderate heat ablation at 300 °C in argon and chemical reduction of GO (CRGO) using a green reagent. They synthesized GO using Hummer's method with pH neutralization and achieved a yield of $\approx 33\%$. At the end

of the process, a bottle of 1 wt% aquatic amber solution was collected and stored and a fraction of the aquatic GO solution was then freeze dried at 100 °C for 48 h. Compared with chemical treatment, the reductive chemical treatment generated additional flaws by fragmenting the platelets.^[46] Besides, Liu et al. proposed a unique flame-induced synthesis method for the mass manufacturing of graphene, which is highly desirable due to its excellent properties and cost-effectiveness. In the process, 3g of GO powder was prepared using Hummer's method and then thoroughly combined with surplus ethanol to produce swollen GO fine particles. The swollen GO fine particles were then lit with a match under stable conditions, with a cylindrical metal net placed above the flame to collect the reduced GO (rGO) powder. Degradation of the functional groups containing oxygen results in graphene-like sheets with random stacking, as shown in field emission scanning electron microscopy (FESEM) micrographs (Figure 3a) of rGO. As stated by the author, van der Waals' cohesiveness between graphene's disorganized stacking at its area boundaries may reduce the material's overall energy. Nevertheless, the author overlooked the limitation on the quantity of energy that could be reduced by disorderly stacking. Nevertheless, still, it is considered an ecofriendly approach, indicating the possibility of adjusting the surface composition of excellent graphene materials for technological applications (i.e., sensors and transistors).^[47]

Jiang et al. further discussed that the high temperature caused a significant decrease and quick expansion of GO during flame treatment, resulting in highly porous structures and randomly dispersed microcracks and microslits. GO paper's usually smooth surface is marred by creases and folds caused by an irregular stacking of the GO layer upon filtration (Figure 3b). Since the GO paper would burn if subjected to a flame for too long, the treatment time was kept to a minimum by performing the process in an ambient setting and is often very short. The tidy and uninterrupted GO paper was transformed into a chapped film, as shown in the

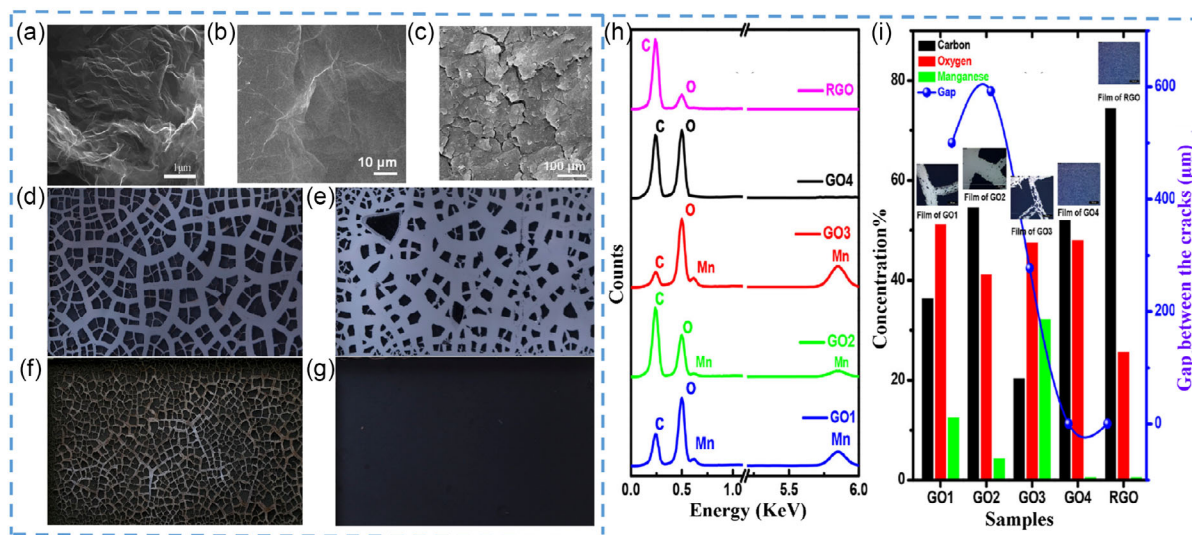


Figure 3. a) SEM micrographs of rGO. Reproduced with permission.^[47] Copyright 2014, Royal Society of Chemistry. b) SEM image of GO paper. Reproduced with permission under the terms of the Creative Commons CC BY license.^[48] Copyright 2015, the Authors. Published by Springer Nature. Digital photograph of dried GO: d) GO1; e) GO2; f) GO3; g) GO4; h) EDS data for all GO1–GO4 and rGO film; i) element composition, fracture spacing relative to GO1–GO4 and rGO film sensor. Reproduced with permission.^[49] Copyright 2023, American Chemical Society.

scanning electron microscopy (SEM) micrographs (Figure 3c) of flame-reduced GO (FR-GO) with many cavities and breaks visible throughout the sample. The quick elimination of oxidized CGs (OCGs) due to the heat impact may have contributed to the creation of this morphology. The author, however, did not provide any practical suggestions on how to prevent cracks and flaws.^[48]

Crack development and methods for preventing fractures in graphene production were reported by Vishwanath and co-workers. Rearrangement and sample size reduction occurred due to a capillary force acting on the molecules during solvent evaporation. At the same time, there was a frictional force between both the substrate and the GO. Cracks (macro- to micrometer scale) develop if intermolecular forces are too weak to prevent movement. When the solvent evaporated, it caused stress via interfacial tension, priming axial tensile force in the film plane. Also, cracks formed and spread throughout the GO film when tensile stress developed inside the sample is higher than the rigidity of the sample. Increases in both the tensile stress and the solvent's partial vapor pressure led to wider cracks in the first place because the creation of larger cracks can only dissipate such large amounts of tension. If the tensile stress was not completely alleviated when the major fractures formed, then secondary cracks appear. However, the drying technique, the substrate quality, and the suspension chemistry might be manipulated to prevent fractures.

The digital photograph shows various GO substrates with cracks (Figure 3d–f) and without cracks (Figure 3g). GO₄ differs by revealing the creation of a multilayered GO film that is homogeneous and free of cracks. Energy-dispersive spectroscopy (EDS) study reveals that the oxygen concentration of (GO precursor GO_(1–4)) GO₁ is more than all other instances, but rGO has a more significant amount of carbon. However, GO₄ has about the same amount of carbon and oxygen (Figure 3h). It is proven that the GO₂ sample has a more significant gap than the other samples. Hence, GO₄ and rGO exhibit a continuous layer since the stacked GO films have no gaps or breaks (Figure 3i). The comparisons revealed that a nearly equivalent combination of oxygen and carbon atoms in GO₄ results in the growth of a GO film without cracks. A reduction phenomenon of oxygen in the EDS curves of rGO demonstrates the successful thermal reduction of GO. The author primarily discussed the amount of carbon and oxygen, confirming that an equal quantity of carbon and oxygen inside the specimen may lead to generating a GO film without cracks.^[49]

Further, Wang et al. considered the toxicity of graphene sheets and their compatibility with living things to determine their potential application in the biomedical field. They created an rGO film utilizing an enhanced thermal decomposition method, that is, step-by-step heating and cooling of the GO hydrosol from ambient temperature to elevated temperatures for 12 h. Later, after cooling to ambient temperature, a thin black film with a shinier and less crumpled structure was formed. Cytotoxicity tests demonstrated the biocompatibility of rGO films. In general, the GO layer is composed of a combination of sp^2 and sp^3 -bonded carbon atoms along with numerous oxygen-containing molecules. After step-by-step annealing, the groups that contained oxygen were eliminated from GO which makes GO to be deformed.^[50] The process enhances the transformation of disorganized sp^2 bond carbon atoms and sp^3 hybridized structural flaws into an organized framework with a significantly higher

concentration of sp^2 hybridized carbon, resulting in a deformed or permeable structure. However, the optimal heating and cooling rates for the structural change and the effects of varying heating and cooling rates remain unclear. Additionally, Sengupta et al. considered the benefits of TRGO, resulting in substantial weight reduction and volume expansion. They presented that the quality of rGO depends on the amount of temperature required at the start for volume expansion amid the thermal decomposition of GO. They used a modified Tour's approach for 7 days to synthesize GO with pure H₂SO₄ and HNO₃ with pretreated graphite powder. In an ice bath, undiluted H₂SO₄ and 30 mL of H₃PO₄ were gradually added to intercalated graphite and KMnO₄ at ambient temperature, which resulted in a ruddy brown solution with continuous stirring and heating to 50 °C for 15 h. Subsequently, after stirring for 2 h, the mixture with water was left for several hours. Next, the acid KMnO₄-dense liquid state was separated 2–3 times for GO filtration. The brown GO cake was cooked overnight at 55 °C to obtain GO. It was observed that Tour's approach yielded 7.5% more GO and required 40% less acid and 33% less KMnO₄. The optimum temperature was 350 °C, leading to the greatest decrease in GO, carbon-to-oxygen ratio, and lattice defects. However, the author overlooked addressing the possible mechanisms behind the highest GO reductions with lesser lattice flaws.^[51] Klemeyer et al. described geometry's influence on GO solids' thermal degassing behavior. A GO foam (Figure 4a) was prepared by freeze drying an aqueous GO dispersion which efficiently generates a GO foam. Under identical heating conditions, denser GO solids had a higher reduction rate, lower peak degassing temperature, emitted more gases, and produced rGO with a higher carbon/oxygen ratio. The SEM image demonstrates the thermal reduction method's operation because freeze drying causes capillary compression. The compact GO mass's inner framework was composed of compressed GO sheets stacked tightly and randomly. Most of the primary explanations for a porous framework are the releases of gases that escape from the system during the heating process. The foam's porous nature is more conducive to rapid gas discharge than the solid's dense packing. In contrast, it consisted of an unstructured porous structure in which GO layers are faintly overlapping and connected without a thermally reduced technique.^[52]

Unless high temperatures and/or a carbonaceous environment are used to promote the appropriate rearrangement and/or accumulation of carbon atoms during thermal decomposition or simple annealing, it is quite improbable that excellent graphene will be obtained. Nonetheless, certain knowledge gaps remain about molecular oxygen removal during GO reduction. In addition, bandgap manipulation of rGO is also essential for organic nanoelectric global applications, which require further investigation of the processes and surface remodeling activities through physical adsorption and chemical modification.

2.2. Aerosol Spray Method

Another method to obtain CGO is aerosol processing.^[53–55] This method is simple and ecofriendly when it starts with an aqueous solution, has a quick processing time, and applies to continuous production.^[56] Several ways to avoid aggregation in solution have

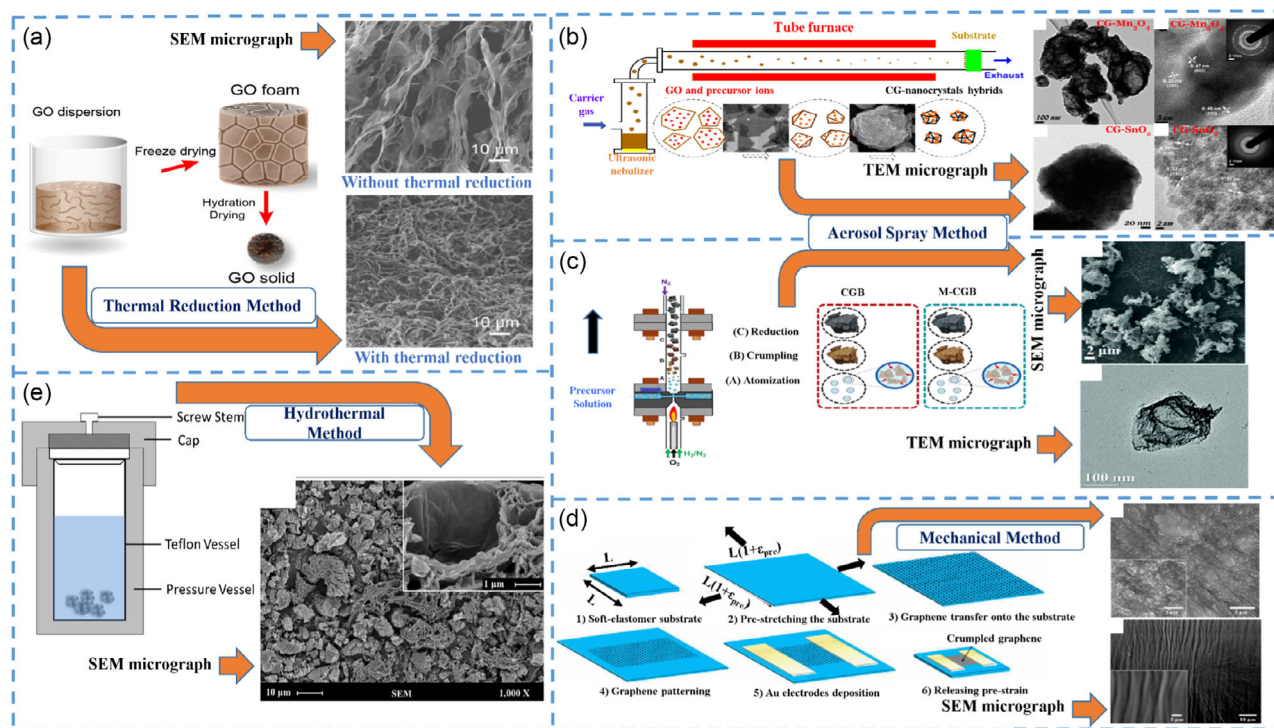


Figure 4. a) Schematic illustrations: synthesis of GO model systems with SEM micrograph. Reproduced with permission under the terms of the Creative Commons CC BY license.^[52] Copyright 2021, the Authors. Published by American Chemical Society. b) Schematics of aerosolization/high-temperature prompt GO crumpling with nanocrystal formation showing in TEM micrograph. Reproduced with permission.^[62] Copyright 2012, American Chemical Society. c) Flame-based method for the fabrication of crumpled, reduced GO nanocomposites with SEM and TEM micrograph. Reproduced with permission.^[73] Copyright 2019, Royal Society of Chemistry. d) The six-step fabrication process flow of a stretchable CG gas sensor produced from mechanical method with SEM micrograph. Reproduced with permission.^[82] Copyright 2019, IEEE. e) Schematic of standard hydrothermal technique apparatus SEM micrograph producing heat-treated graphene oxide sheet. Reproduced with permission under the terms of the Creative Commons CC BY license.^[95] Copyright 2019, the Authors. Published by MDPI. Reproduced with permission.^[162] Copyright 2022, Elsevier.

been explored, including sheet size reduction, solvent-graphene interaction modification, and dispersion agents. Luo et al. demonstrated how ultrasonic atomizer-nebulized GO dispersions in the water of varying concentrations generated aerosol droplets transported by N_2 gas at 1 L min^{-1} via a horizontally placed tube furnace. They observed that the surface of crumpled grapheme was continuous and more prominent than conventional flat sheets processed under the same circumstances.^[12]

Similarly, Xiaofei et al. aerosolized GO in an aqueous solution and was quickly dried to create nanopaper-like crumpled sheets. They preoxidized GO using a modified Hummers' technique. They eliminated salt byproducts from the GO sample using acid-acetone wash. According to the authors' predictions of confinement force, CG sheets' formation is the result of capillary force and fast solvent depletion.^[57] Guo et al. and Jo et al. studied different graphene structures for gas storage, separation, catalysis, and supercapacitor applications. They determined the influence of the morphological structure on the electrochemical characteristics of CG balls (CGBs) with varying particle sizes and pore diameters.^[58,59] In a conventional spraying procedure, the initial solution was atomized (or nebulized) by an atomizer (or nebulizer) to create tiny droplets in a heated furnace, where solvent evaporation, solute diffusion, precipitation, drying, pyrolysis, and sintering aided in particle formation.^[60] The

ultrasonication method mixes a GO powder to produce a GO suspension. An ultrasonic nebulizer helps create aerosol particles from the suspension. Argon later transports H_2 , N_2 , or other carrier gases into a warmed horizontal tube furnace at $600\text{--}700^\circ\text{C}$, where GO sheets are rapidly dried and crumpled. The average droplet size created by the ultrasonic nebulizer shown in Figure 4b was $\approx 1.7 \text{ mm}$, which caused the solvents to evaporate in the tube furnace quickly and compressed GO sheets into submicrometer-sized balls. The folding structure in the transmission electron microscopy (TEM) image shows the intertwined carbon lattice structures. The CG- Mn_3O_4 and CG- SnO_2 combinations have hollow structures with the same nanocrystal patterns inside and outside the CG balls. This makes sense since the precursor ions are held on both sides of the GO layers before the aerosols are released. The author states that structural categorization holds great potential due to the following benefits. The TEM image reveals excellent metallic oxide dispersion in CG; hence, future CG-based hybrids with diverse nanocrystals comprising metals and metal oxides are preferred. The precursor's concentration, dispersing aerosol temperature, and precursor source may be adjusted to tailor nanocrystal size and shape to a given application. As a result of plastic deformations, CG structure is stabilized by locally folded, layered ridges, capable of withstanding redissipation in a liquid solvent without the structure

unfolding or collapsing.^[61,62] The crumpled structure has a larger specific surface area than frequently used flat GO/graphene sheets. Therefore, considering that a hollow/open framework allows nanocrystals to assemble on both the internal and external surfaces of the CG, it is anticipated that this novel hybrid structure has the possibility of CG-based design modifications in functional sensing applications. In addition, Cheng et al. provide a spray freezing technique for rGO/MoS₂ composites used in energy storage applications. After further dehydration and thermal processing of GO with (NH₄)₂MoS₄ solution, a 3D structure comprising rGO and tiny MoS₂ layers were formed. Nevertheless, the dispersion of rGO was improper (agglomeration), which is not addressed by the author, yet they claim that the composite derived from their synthesis technique displays greater dissemination than the solid-liquid synthesis approach in their study.^[63]

Some aerosol routes have shown that the shape of the crumpled spheres might not be significantly affected by the furnace temperature once the process is done, indicating crumpled spheres were already formed before they entered the hot furnace.^[64] Guo et al. investigated the response of GO nanosheets to water by folding and unfolding in the aqueous phase. The GO nanosheets were systematically grouped into complex, multilayered structures through surface anchoring, flow, and microconfinement.^[65] Chen et al. explained that monolayer GO could be cosuspended in dilute aqueous phases for applications that require the isolation of nanoscale elements from the environment or biological substances.^[66] Carraro et al. evaluated a novel aerosol synthesis technique that enabled the concurrent doping and crumpling of GO. They generated N- and N-, S-doped graphene structures with 2 g of graphene. Then, 6 g of KMnO₄ mixture in an ice–water–acetone bath was heated for 2 h at 35 °C. Next, 100 mL of deionized (DI) water was slowly heated to ≈98 °C and maintained for 15–20 min. Stirring and sonication for 30 min yielded a yellow product dispersed in GO with a pH of 5.5.^[67] Choi et al. prepared CG–MoO₂ composites using Mo ions in a stable GO colloidal solution and spray pyrolysis.^[68] CG and CGBs made of various 3D graphene–metal oxide composites are preferred for multiple applications with exceptional graphene performance.^[69–72] Mohammadi et al. developed a flame-based method for the continuous one-step manufacturing of 3D graphene nanostructures. The capillary compression prompted by the rapid evaporation of droplets, as shown in Figure 4c, led GO sheets to self-assemble into crumpled balls. The resulting CGBs decreased the oxygen-containing groups in GO due to excess hydrogen at high temperatures. Aerosol droplets containing metal alloy-coated reduced CGBs included GO and metal precursor ions. Metal ions in the solution might be adsorbed onto GO sheets via electrostatic interactions. The SEM and TEM images demonstrate the formation of a deformed structure and metal deposition immediately onto the grid of particles from the product aerosol. Metallic nanoparticles (NPs) have entirely covered the CGB and exhibit little agglomeration compared to typical aerosol products. The structure of the metal design suggests that metal NPs form from metal ions adsorbing on the sheets of GO in the precursor fluid. If metal NPs are generated in a gaseous state, we anticipate these individuals to form branched or irregular aggregates rather than uniformly dispersed on the GO surface.^[73] Koirala et al. reported that the flame aerosol technique is suitable for producing various (3D composite)

heterogeneous catalysts because it allows the rapid and scalable synthesis of functional NPs.^[74]

While this is the most common technique for preparing CG, other possibilities, such as adjusting the molecular formula, developing a cheap design for mass manufacturing, and codoping with other metallic elements and elements from rare earth, are still open. Whereas the CG–Pt composite^[75] had the maximum current flow and high sensitivity, it may also be codoped with other noble metals to improve their electron transfer capabilities. Researchers have observed that CG affects biological viability, including an interaction between normal tissue and GO. Nevertheless, the mechanism of this connection is yet to be investigated to get optimal sensing capabilities.

The efficacy of a sensor varies with the synthesis of crumpled graphene and the fabrication procedure, as shown in the comparison table. The hydrothermal method is recommended to determine the best technique in terms of cost and ease of preparation of CG, as its temperature requirement is less than 200 °C and requires a smaller fabrication setup than the thermal reduction and aerosol spray methods. In addition, the mechanical synthesis of crumpled graphene is the most cost-effective method, but it requires more design and experimental apparatus, so it may not be suitable for bulk production.

2.3. Mechanical Method

Concentrating bending and stretching energy in the stretching grooves is essential to crumple formation. Flattening a crumpled paper ball reveals a complex network, which indicates the uneven distribution of elastic energy in the structure. This behavior is attributed to fixed 2D connectivity, which has two significant effects. First, shear stiffness, bending, and extension produce stress concentrations in thin ridges.^[76,77] Second, the importance of the object's "self-avoidance," or the way the parts repel each other, sharply increases as the number of its internal components grows. In other words, self-avoidance is thus more critical for sheets than continuous polymer chains. They also observed that significant deformation of a film of solid mass usually results in a crumpled condition with sharp points with high curvature.^[78] One potential reason for the deformation behavior is that the boundary condition must be maintained precisely throughout the loading operation. For instance, the stresses in the facets and the effective operating conditions for every ridge may vary as a function of the forces operating at the vertices. In order to know how crumpled sheets will react elastically, we need to apply load in the direction opposite to the ridge's weakest link. Because of its strong symmetry, the selected load may lack a component in the ridge's weakest direction. Thus, the response rigidity of the ridge to such a force is qualitatively larger than its response stiffness to a generic load.

Vliegthart et al. simulated a model of 3D structures bending and stretching stiffness at a coarse scale of self-avoiding elastic sheets.^[79] Besides, Zang et al. proposed unique CG–paper electrodes with outstanding stretchability, specific capacitance, and reliability. They fabricated nanoporous graphene paper with a high packing density. Also, they described that the elastomeric sheets' prestretching relaxed, and the graphene sheet's lateral dimensions dropped proportionally.^[80] Equally, Kang et al.

proposed a new method for fabricating plasmonic structures that can be changed mechanically and have strong resonance. Their calculations demonstrated that physically changing CG structures allowed them to tune plasmonic resonance across a broad spectral range.^[81] Shao et al. suggested an improved sensing mechanism model on the CG gas sensor fabricated with a 200% stretching ability. Figure 4d shows the six-step synthesis process of the CG gas sensor, which is a biaxially stretched elastomer substrate. Due to mismatch in the modulus of elasticity between graphene and the substrate, the SEM images depict the spontaneous creation of graphene wrinkling following the release of the prestrain. The degree of resistance significantly altered when microscale wrinkles appeared due to exerted pressure in one direction and static pressure in the other. It has been shown that graphene resistance relies heavily on the imposed tensile strain. The exceptionally high sensitivity to the stress of the compressed graphene sheets indicates the material's great potential for use in strain measurement devices. Additional experiments are required to investigate the applicability of deformed graphene sheets in flexible electronic devices.^[82]

Nonetheless, knowing how crumpled graphene reacts to mechanical stress is crucial for integrating them with other materials in various applications. To generalize single-fold dynamics toward the mechanical response of macroscopic scale clusters is necessary to investigate each nanometer-scale crumple's mechanical characteristics and force sensitivity. Furthermore, strategically designing materials for improved energy and materials preservation, energy dissipation, and tribo modification requires a deep comprehension of mechanical characteristics at the nanoscale.

2.4. Hydrothermal Method

Chemical vapor deposition (CVD) requires a high reaction temperature, which has limited its applications. Thus, a simple method to produce a significant amount of graphene is needed, that is, the hydrothermal reaction, which leads to a high product yield at a low reaction temperature.^[83,84] Mei et al. synthesized rGO by reducing GO using the hydrothermal method at various temperatures. They thoroughly studied the effect of the reaction temperature on the interlayer distance and intensity ratio. The as-synthesized GO (50 mg) was sonicated for 30 min and then dispersed in ethylene glycol, resulting in a brown colloidal solution. The sample was kept in a sealed 50 mL Teflon-lined autoclave for 16 h at various temperatures. The solids were separated by filtration and then rinsed using acetone and distilled water. The samples were finally cured in a vacuum chamber at 65 °C for 24 h. Their findings show that reaction temperature affects GO reduction, although the mechanism and optimal temperature and reagents for reduction and oxidation are still unknown.^[85] Sieradzka et al., who studied the effect of oxidation and reduction factors on rGO, noted that graphene oxides with more oxygen molecules in their structure resulted when the reactants were combined for an hour before the oxidation began. Meanwhile, the shape of the final product is heavily influenced by the data collected during thermal reduction and by the amount of water present in the reduced samples. When rGO was in the shape of a slurry having 70% water, a homogenous powder was produced.

In addition, microexplosions conducted at higher temperatures were also used to remove oxygen functional groups from materials containing up to 70% water. They also described how the microexplosion initiated at temperatures around 200 °C led to a decrease in size by a factor of 10, causing a dramatic shift in the underlying structure.^[86]

Researchers are witnessing the development of cutting-edge methods for preparing nanoscale materials. Among these, the microwave hydrothermal approach uses the microwave hydrothermal concept for heating but differs from conventional hydrothermal synthesis. Microwave hydrothermal technology combines the best features of hydrothermal and microwave heating systems. Because the microwave hydrothermal approach shows a higher heating rate than the conventional hydrothermal method, it can rapidly manufacture NPs with consistent size distributions and shapes.^[87–90] This approach is inexpensive and environmentally benign, with a short processing time. The primary advantage of this approach is the effective energy transmission and quick volumetric heating. This process shows a limited but technologically advanced system with controlled heating.^[91] In addition, the nanocomposites produced by the microwave-assisted hydrothermal approach have excellent properties such as high output, simple production methods, and low-energy consumption.^[92,93] Police et al. showed that hydrothermally grown TiO₂ nanotubes over rGO layers exhibited ≈ 12.9 times more photocatalytic selectivity toward H₂ production than commercial TiO₂ powder (P25). An alkaline hydrothermal process produced TiO₂ nanotubes with varying GO content. To obtain homogeneous GO dispersions, 0.5%, 1%, 2%, 5%, and 10% (wt%) GO were stirred continuously in DI water for 1 h.

TiO₂ (P25) was added gently to a GO suspension during stirring. A further hour was used to ensure that the solution was fully blended. Then, NaOH mixed solution was poured into an autoclave lined with Teflon for 24 h at 130 °C. The sample obtained (gray gel) from a Teflon mould was stirred for 24 h after washing with DI water and 0.1 M HCl solution at ambient temperature. The finished product was rinsed several times using DI water, centrifuged, dried, and then annealed at 300 °C for 4 h. They found that photocatalytic efficiency is highest at 5 wt% rGO, improving electrolyte mobility and electrochemical performance.^[94]

Hydrothermal synthesis provides many opportunities for the researcher to determine the optimal parameters for a specific study, despite the difficulties inherent in its use for mass production and to keep up industry-level product quality. Oxides and oxygenated salts comprise the bulk of materials processed via the hydrothermal route. Therefore, enhancing the fabrication of materials using hydrothermal techniques requires research into the physicochemical features of hydrothermal devices (Figure 4e), solvents, and mineralizers and an understanding of chemical reaction mechanisms during hydrothermal processes. The SEM micrograph indicates that significant cracks and ruptures are created due to the gas pressure resulting from the breakdown of oxygen-related compounds on the graphene layers, thereby decreasing the van der Waals attraction between neighboring graphene sheets during the heating process (200 °C). Furthermore, by mixing graphene sheets with various materials, the hydrothermal method might decrease certain O₂-related groups on GO and result in the restoration of a π -conjugated structure. Hydrothermal treatment requires

relatively less heat and is regarded as one of the most affordable techniques for producing graphene with a crumpled design.^[95] Hydrothermal techniques' practical scope and theoretical foundation seem to benefit from the expanding fields of current materials science and engineering research. Hydrothermal technologies are expected to gain popularity owing to the trend toward interdisciplinary collaboration in which they are combined with other methodologies.

2.5. Control Crumpling

Wrinkle/crumple graphene's reversible form is necessary to control since its deformation could significantly affect the properties and functionality of graphene-based devices.^[96,97] Thermal expansion and capillary compression could cause graphene to wrinkle and crumple. However, the processes of reversible crumpled formation of graphene sheets remain poorly understood.^[98–100] Zang et al. proposed a straightforward method of controlling the reversible crumpling of graphene sheets, which generated unique super hydrophobic, transparent, and wettability- and transmittance-tunable conductive coatings and electrodes. They demonstrated the self-organization of graphene sheets into hierarchical patterns resembling superhydrophobic leaves. Concentrating on the formation of wrinkled and

delaminated buckles, the first wavelength (λ)-dependent wrinkle was created when the deformation in graphene reached a critical level.

$$\lambda = 2\pi t \left[\frac{E}{12\Lambda\mu(1-\nu^2)} \right]^{1/3} \quad (1)$$

where E , ν , μ , and t are the elastic moduli and Poisson's ratio, shear modulus, and thickness of the film. $\Lambda = (1 + (1 + \epsilon)^3) / 2(1 + \epsilon)$, where ϵ is the prestrain of the substrate. They demonstrated that polished graphene/PDMS specimens could be stamped on a biaxial prestretched acrylic elastomer film. The author outlined several potential novel research avenues, including rigorous and statistical studies of how graphene crumpling affects its electrical and electrochemical characteristics and the robustness of graphene/polymer interactions. The crumpled graphene's nonuniform structure is severely distorted and microscopically structured, possibly leading to further novel features and functionalities. In addition, novel graphene-based systems with innovative tunability and flexibility may be developed by manipulating graphene's tiny patterns with a simple macroscopic device, thereby rendering nanoscale mechanics observable at the macroscale.^[101]

In addition, Thomas et al. showed that GO films allow high optical transmission compared to graphene. **Figure 5a** shows

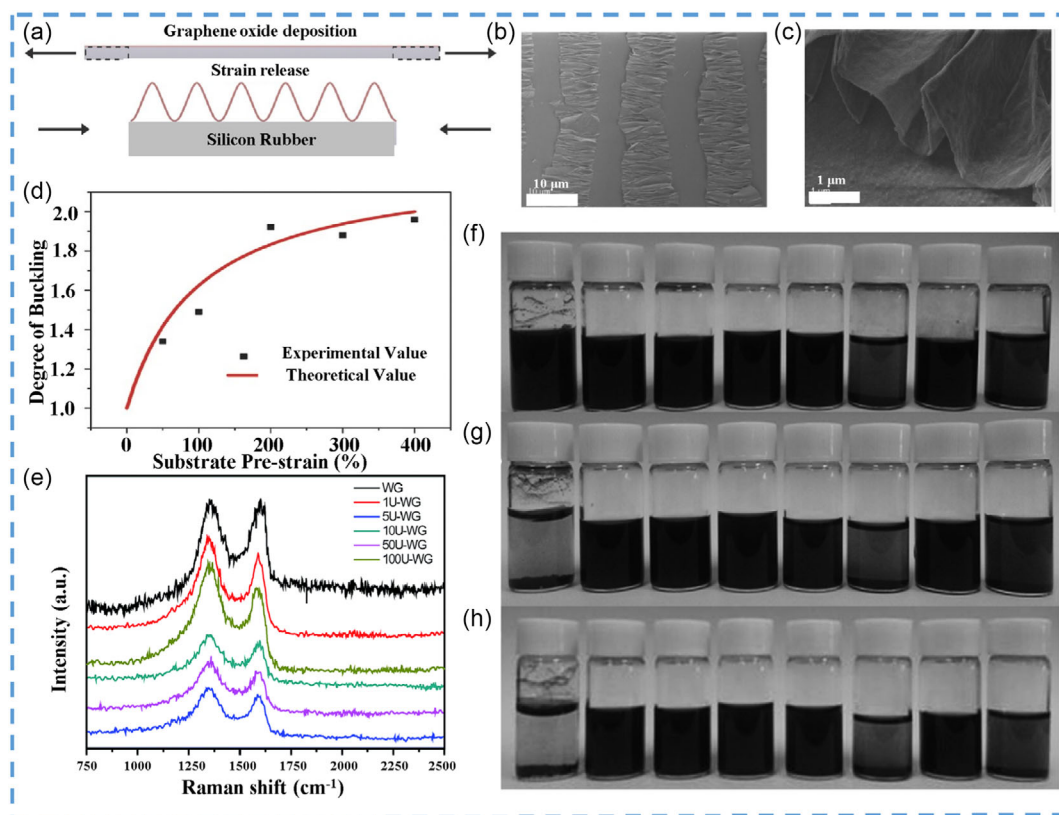


Figure 5. a) Schematic illustrating GO formation and strain discharge generating impulsive delamination buckles. b) SEM micrograph representing delamination buckling waveform of GO sheet on the SR substrates under $\approx 400\%$ uniaxial strain release. c) High-resolution SEM micrograph of a single blister with distinct pleats. d) Appropriate statistical predictions of GO film buckling under biaxial compression. Reproduced with permission.^[103] Copyright 2015, Wiley-VCH. e) Raman spectra of different U-WG samples: f) 5U-WG; g) 50U-WG; and h) 100U-WG in different solvents after 0.5 h. Reproduced with permission.^[104] Copyright 2016, Royal Society Chemistry.

a schematic of the impulsive buckle debonding technique. The film was delaminated from the silicone rubber surface in some regions and adhered to the substrate in others. Periodic cracks were apparent and orthogonally directed toward the delaminated blister due to transverse tensile stress induced by uniaxial substrate compression throughout its length, as shown in Figure 5b, c. They observed that reducing the range of modulation of the film's transmittance, fractures, or voids might significantly diminish its opacity. The substrate prestrain significantly influenced a film's overall degree of buckling (d_b) (Figure 5d).

$$d_b = 1 + \frac{(1 + \nu_f)d}{1 + d} \quad (2)$$

where d and ν_f represent prestrain and Poisson's ratio of a film, respectively (for GO $\nu_f = 0.2$).^[102] In addition, applying and releasing the biaxial strains on the silicone rubber substrate is a feasible method to manage the delamination buckling of GO films effectively.^[103]

Preparing graphene with an intentionally directed shape and open structure remains a challenge. However, Tang et al. focused on crumpled graphene ball (CGB) and their confined spherical shape to use the restricted interior surface. In addition, the urea content significantly influenced the N-doped CG (NCG). Figure 5e shows that integrating nitrogen atoms into the carbon network caused more defect sites. Raman spectra further demonstrate that the weight ratio of urea to graphene (5:1, U-CG) causes urea to saturate all interior graphene surfaces.

However, a low quantity of urea had negligible effects on graphene crumpling. As urea concentration increases, the NCG progressively transforms from a spherical to an open structure. Figure 5f–h shows that the shape of graphene significantly influences its dispersibility and aggregation resistance. The sample 5U-WG exhibited outstanding dispersibility in almost all solvents. The sample (50 and 100) U-WG performed poorly in water and benzene. The solution mass determined the capillary force, which shows inverse relation to the particle size of the crumpled graphene. The confinement force was assumed to be constant regardless of particle diameter or mass concentrations at the predetermined heat treatment temperature. Thereby, the crumple content was mainly affected by the thickness of the GO/urea composite, which in turn affected the resistance capacity to the confined force. Precipitated material on the internal layer with thickness t can be determined using droplet size as a variable

$$t = V \cdot A^{-1} \quad (3)$$

$$V = \gamma \cdot m \cdot \rho^{-1} \quad (4)$$

$$m = \frac{4}{3} \pi (R^3 - r^3) C \quad (5)$$

$$t = \gamma \cdot C (R^3 - r^3) (3\rho r^2)^{-1} \quad (6)$$

V , m , ρ , and C equal urea's volume, theoretical mass, density, and concentration, respectively. A indicates the droplet's surface area; $\gamma = (\text{mass of urea}/\text{mass of GO})$; R = initial radius, r = radius of reduced graphene ball. Equation (3)–(6) show that controlling the radius of a crumpled ball can manipulate the thickness of the sample, which causes the control of the macroscopic property of the sample. Contrastingly, it also can be observed that graphene

with slight wrinkling can only be dispersed in certain specialized solvents.^[104]

Improved energy storage in supercapacitors, sensors, and actuators may be possible because of wrinkles' increased surface area. Nonetheless, while preparing electronics, it is essential to minimize the appearance of wrinkles. For instance, when evaluated, the sheet resistance values for wrinkled films varied widely. For optical applications, wrinkles are a problem because their roughness and inhomogeneity cause light scattering. Although several studies have attempted to develop the process underlying the control wrinkle/crumpling of graphene, more precise ideas are required to determine the connection between controlled crumpling and the characteristic variability of graphene.

3. Sensor Application

3.1. Biosensor

The crumpling of graphene improves mechanical stability and addresses the balance between sensitivity and processability in sensor applications. Crumples in substrates' conductive layers increase the contact surface and current channels, improving sensor responsiveness and performance.^[105–108] Graphene-based gas sensors can adsorb individual gas molecules with greater sensitivity. Gas sensor arrays enhance a catchment area,^[109] are sensitive to short-term exposures, and detect traces of toxic gases.^[110,111]

Leem et al. incorporated structural 3D into Raman enhancements and developed a mechanically driven manufacturing technique for 3D hybrid CG–Au NP nanoplasmonic structures. The researchers found that a 3D hybrid CG–Au NP nanoplasmonic structure could be fabricated from thermally induced shape memory polymers. **Figure 6a,b** shows the mechanical construction process of the 3D hybrid CG–Au NP structure for high-sensitivity applications. The scanning electron microscopy image of flat and hybrid CG–Au NP structures shown in **Figure 6c,d** explains the uniform distribution of Au NP onto the CG surface. Further, in comparison to two distinctive Raman peaks (CG/flat graphene–Au NPs substrate), **Figure 6e** demonstrates that a CG–Au NP substrate has a higher amplitude peak than a flat substrate. The limit of detection (LD) and the Raman enhancement factor show that the crumpled substrate was at least 10 times greater than the flat substrate. Two parameters work together to provide the crumpled graphene–Au NPs hybrid system with its enhanced intensity and detection sensitivity: 1) greater concentration of nanoplasmonic “hot spots” and 2) closer vicinity between both the NPs to generate “hot spots” with a greater amplification. Densification of Au NP in a particular 2D region takes place owing to the debonding of graphene whenever the PS substrates deflect under a compression load. The right panels of **Figure 6f** show relatively low-field augmentation ≈ 2.5 and B–B' curve of flat graphene–AuNPs composite, while the left panels demonstrate incident electromagnetic enhancement by ≈ 5.5 and the line narrative of electromagnetic wave amplitude all along the A–A' line of crumpled graphene–Au NPs structure. As the Raman intensity is related to the fourth power of the incident electromagnetic enhancement, a 3D structure would be projected to display a Raman enhancement factor up to 24.29 times

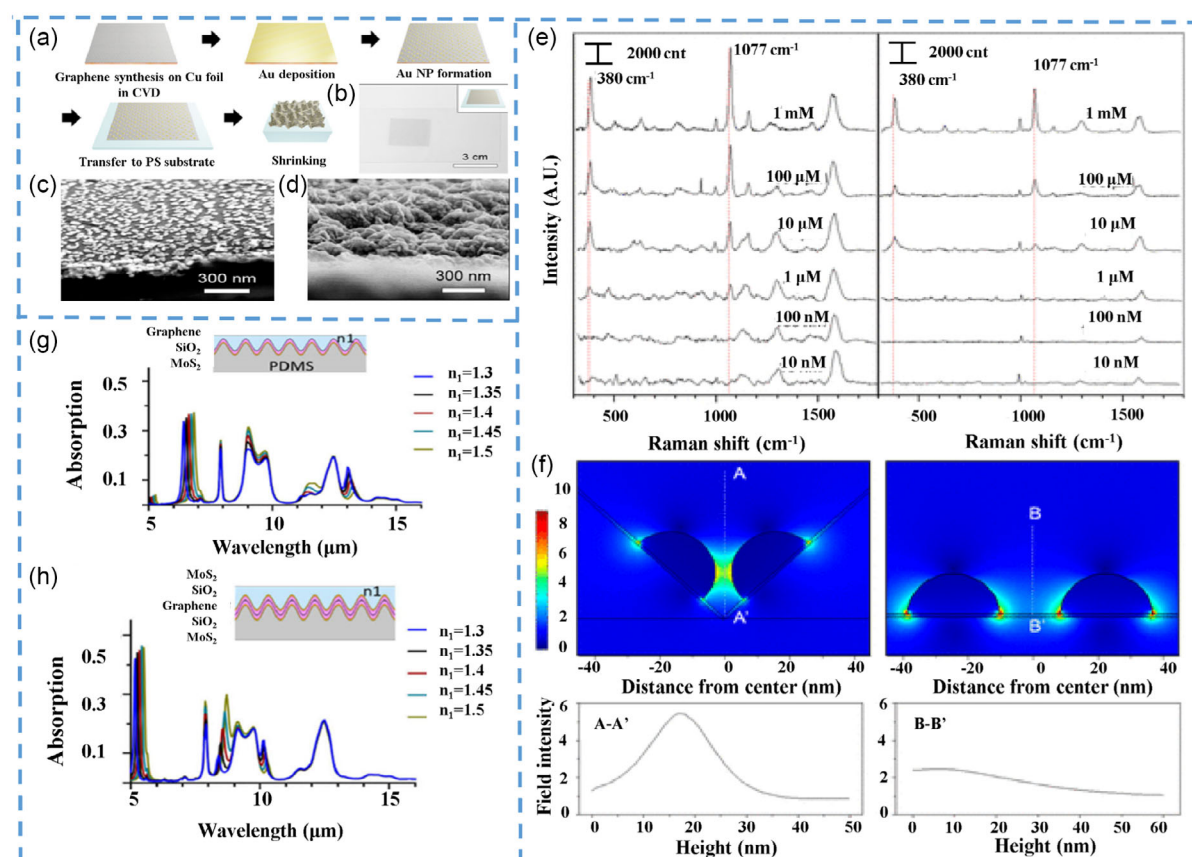


Figure 6. a) The blended crumpled CG–Au NP structure put onto a PS substrate reduced by heating it over its glass transition temperature to create 3D hybrid structures; b) SEM micrograph of the hybrid CG–Au NP plasmons at nanometer-scale structure on PS substrate c) before and d) after shrinking. e) Surface-enhanced Raman spectroscopy of thin graphene–AuNP substrates showing a comparison of plasmonic resonance shifts of various graphene composite heterostructures. f) COMSOL simulation of CG (left) and flat (right) structure. Reproduced with permission.^[112] Copyright 2015, American Chemical Society. g) Standard optical absorption spectrum of a CG/MoS₂/SiO₂/g. h) Crumpled MoS₂/SiO₂/graphene/SiO₂/MoS₂ structures supported on corrugated polydimethylsiloxane substrate. Reproduced with permission under the terms of the Optica Publishing Group Open Access Publishing Agreement.^[118] Copyright 2021, the Authors. Published by The Optical Society.

larger than that of a flat structure.^[112] Transforming nanoscale structure from 2D to 3D increases NP density in a laser-focused volume, enhancing Raman enhancements. Crumpled-graphene nanomaterials with high amounts of pores and readily available surface areas are ultrasensitive in Raman spectra due to the great electrical conductivity and quick transfer of charge of the redox species.^[113–115] This shape significantly decreases the NP gap distance in crumpled graphene structural valleys, which enhances electromagnetic Raman intensity. Several research gaps, such as the author's seemingly arbitrary choice of target chemical and solvent, are still visible despite the findings' hopeful future implications. As a result, altering the target molecule may somewhat impact the results. Besides, pattern graphene sensors may induce plasmonic resonance,^[116] although pattern design and bandwidth-tunable graphene plasmons are challenging to achieve.^[117] Faramarzi et al. proposed a plasmonic resonance biosensor made of CG and molybdenum disulfide (MoS₂) flakes. Flexible crumpled new 2D materials and 2D material plasmonic heterostructures were used as plasmonic biosensors and compared with the primary plasmon resonance of CG on a PDMS substrate. Figure 6g,h shows that compared with the

PDMS-based graphene sensor, the crumpled MoS₂/SiO₂/graphene heterostructure has improved sensing performance which can be understood by variation in resonance peak. The strong absorbance of the PDMS substrate has a dampening impact on the graphene plasmonic resonances and the responsiveness of the device. Due to the blueshift, the absorbance peaks of PDMS and the plasmon resonance wavelengths are significantly out of tune in the crumpled graphene–MoS₂ heterostructure. In the sensitivity of biosensors, this is crucial because it results in a resonant peak with greater intensity and narrower full-width-at-half-maximum (FWHM). The semiclassical Drude model coupled with the finite element analysis (FEA) helped to find the plasmonic frequency response of a crumpled 2D materials sensor when the index of refraction of an analytical solution changed. The COMSOL Multiphysics-based structural analysis approach efficiently simulates a graphene-integrated device using a thin-film conductive model. The most cross-sectional surface of 2D substances that interact with biomolecules is made possible by the suggested plasmonic sensors with a 3D CG/MoS₂ configuration structure, particularly in the non-uniform areas of the crumpled framework. The author claims

that mechanical remodeling of structural characteristics allows for tuning sensors throughout an extensive spectrum range, which is not well recognized.^[118]

The field-effect transistor (FET)-based diagnosis used for biomolecular sensing platforms with applications in electronic components, including data analyzers and signal transducers, has attracted considerable interest.^[119,120] Biosensor applications require high ionic strength for DNA/RNA detection. Furthermore, this detection potentially replaces hazardous tumor cell biopsy with liquid biopsy in numerous diagnostic applications.^[121] Some articles explain that single-wall carbon nanotube (SWNT)/FET and nanoelectronic sensors based on synthesized 1D and 2D nanomaterials can mitigate the fundamental ionic screening effect by high-sensitivity real-time biological entity recognition.^[122,123] Flat 2D semiconducting materials like MoS₂ show high sensitivity than quasigraphene.^[124] Some modifications, like nanoscale bending of 2D graphene in 3D, may increase sensitivity due to control of a Debye length and strain-induced bandgap widening in graphene channels. Hwang et al. demonstrated the most unorthodox graphene FET-based biosensors that enabled high sensitivity to detect biomolecules with an LD as low as 600 zM on millimeter-scale structures.^[125]

On optimizing CG-based biosensor applications, Jiang et al. reported that optimized CG FET biosensors could detect biomarkers of various sizes with remarkable sensitivity. They also showed that the CG FET biosensor could detect SARS-CoV-2 antigens.^[126] Furthermore, Angulo et al. illustrate that the interleukin 6 (IL-6) protein helps the immune system fight inflammation and bone metabolism. Thus, monitoring IL-6 levels in the blood is extremely important when an infection is present because it can cause harmful effects.^[127] Hwang et al. illustrated IL-6 detection, as shown in **Figure 7a,b**. Because of the significant distance, the flat detector could not identify the IL-6 protein. The CG FET (1 × PBS) can detect attomolar (aM)-level sensitivity. Figure 7c shows that monitoring N-protein in 1 × PBS alters the *I*-*V* curve in CG channels. Even if the author's determination of the isoelectric point of an N-protein (+ve charged) contradicted (−ve charged) previously published findings, the positive charges on the surface may change conductance as the active channel surface attracting the negatively charged areas of N-protein trapped on the surface. Figure 7d shows a noticeable change where the S-protein detection signal was not statistically meaningful and could not be identified due to the enormous diameter of the S-protein antibodies and the reduction in Debye length.^[128] Due to the constrained character of the curve-deformed region,

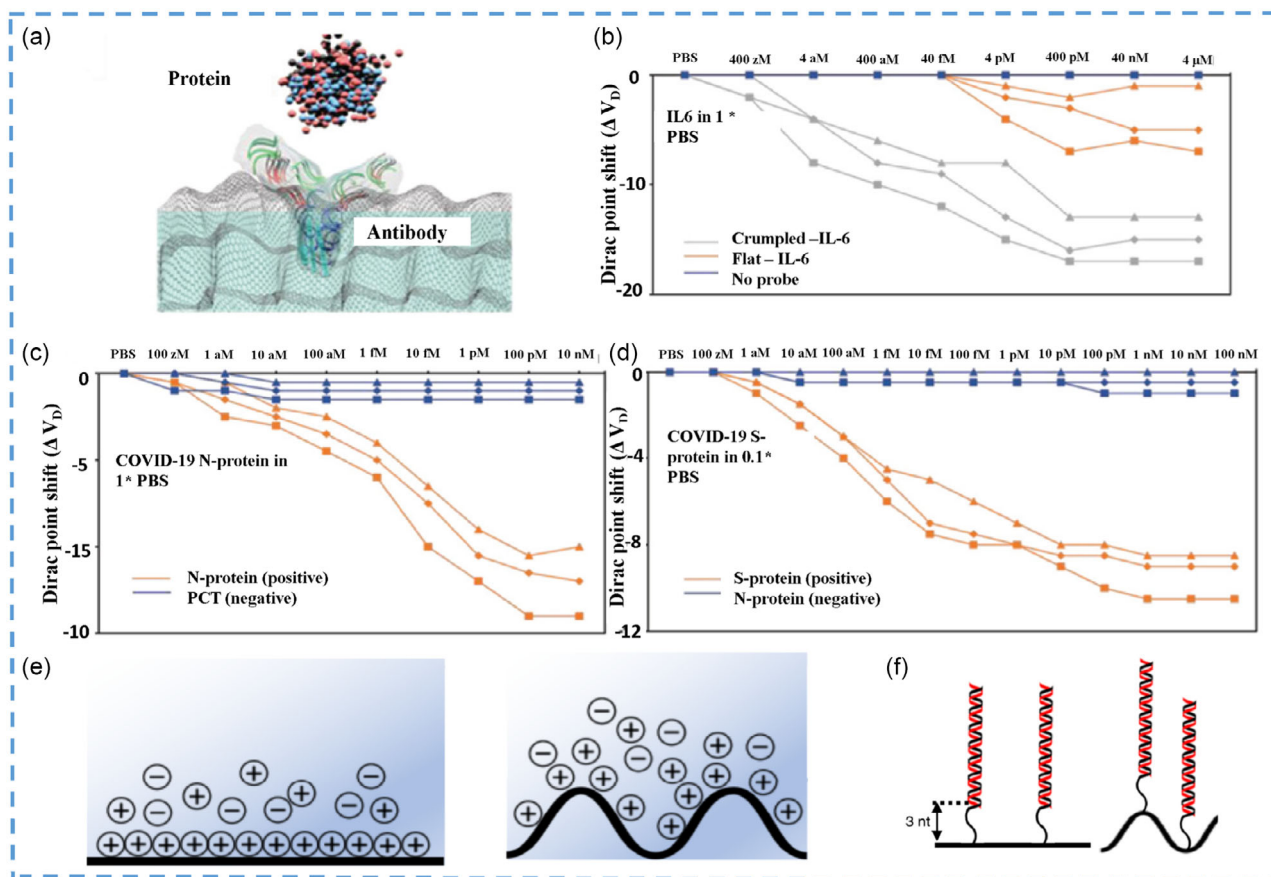


Figure 7. Various FET biosensors: a) Schematic of crumpled graphene protein channels obtained with suitable antibodies; b) FET biosensor Dirac voltage shifts with IL-6 protein diagnosis; c) COVID-19N-protein; d) S-protein. Reproduced with permission.^[128] Copyright 2021, Wiley-VCH. e) Distribution of ion on the surface of flat and crumpled shape and f) Debye length comparison of flat and crumpled graphene. Reproduced with permission under the terms of the Creative Commons CC BY license.^[125] Copyright 2020, the Authors. Published by Springer Nature.

an ionic layer forms at a greater distance from the curved graphene outer layer than in the case of planar graphene. As the frequency of the crumpled form increases, more ions are precluded from the graphene interfaces, as shown in Figure 7e,f. In addition, ion filtration occurs at a substantial distance from the graphene surface.^[125] Moreover, biomolecular detection is improved because fewer detection charges are filtered out in highly crumpled graphene films. Consequently, it is essential to recognize that the effectiveness of twisted graphene might be associated with the crumpling percentage and carefully optimized for higher efficiency.

It is known that the Debye length increases when there are irregular and significant flaws in the structure of twisted graphene, which defines that when a molecule is attached to the surface, the conductance of electricity changes more than it does on flat graphene. A large, confined channel on twisted graphene allows molecular DNA with low ionic screening. Consequently, flawed graphene may decrease the mitigate ion screening of DNA molecules, thereby increasing the sensitivity of biomolecule detection.

Yan et al. emphasized that biaxial CG structures shifted the plasmonic resonance wavelength more than uniaxially standard flat graphene-based devices for a particular refractive index change.^[129] Further, it was observed that because of the high absorption spectrum of PDMS, mainly in the midinfrared region, the substrate's dampening impact altered the graphene plasmon

resonances.^[130,131] Additionally, the biaxial crumpled graphene structure had more robust plasmonic modes, which increased the sensor's sensitivity and figure of merit compared to the uniaxial crumpled structure.^[132] Crumpled graphene outperformed unmodified graphene in biosensor applications. However, innovative and economic applications are not yet fully established. In addition, a different form of graphene sensor is believed to be efficient for drug screening and high-throughput diagnostics.

3.2. Gas Sensor

Graphene is distinguished by its honeycomb-like lattice structure, which exhibits exceptional capabilities for use in specific fields.^[133,134] Recently, researchers have attempted to convert ideal 2D graphene nanosheets into distorted 3D formats.^[135–137] Chen et al. and Ban-Weiss et al. group demonstrated a 3D crumpled bioinspired design NO₂ gas sensor to detect toxic gasses.^[34,138]

Chen et al. used lyophilization to transform 2D flat graphene-based nanosheets into 3D crumpled structures for NO₂ detection. These crumpled nanosheets, used to create gas sensors, strongly react to NO₂ at ambient temperature. Their research proved that 3D crumpled sodium 1-naphthalene sulfonate (NA)-rGO nanosheet has a better option than graphene-based sensors, as shown in Figure 8a. The sensors displayed

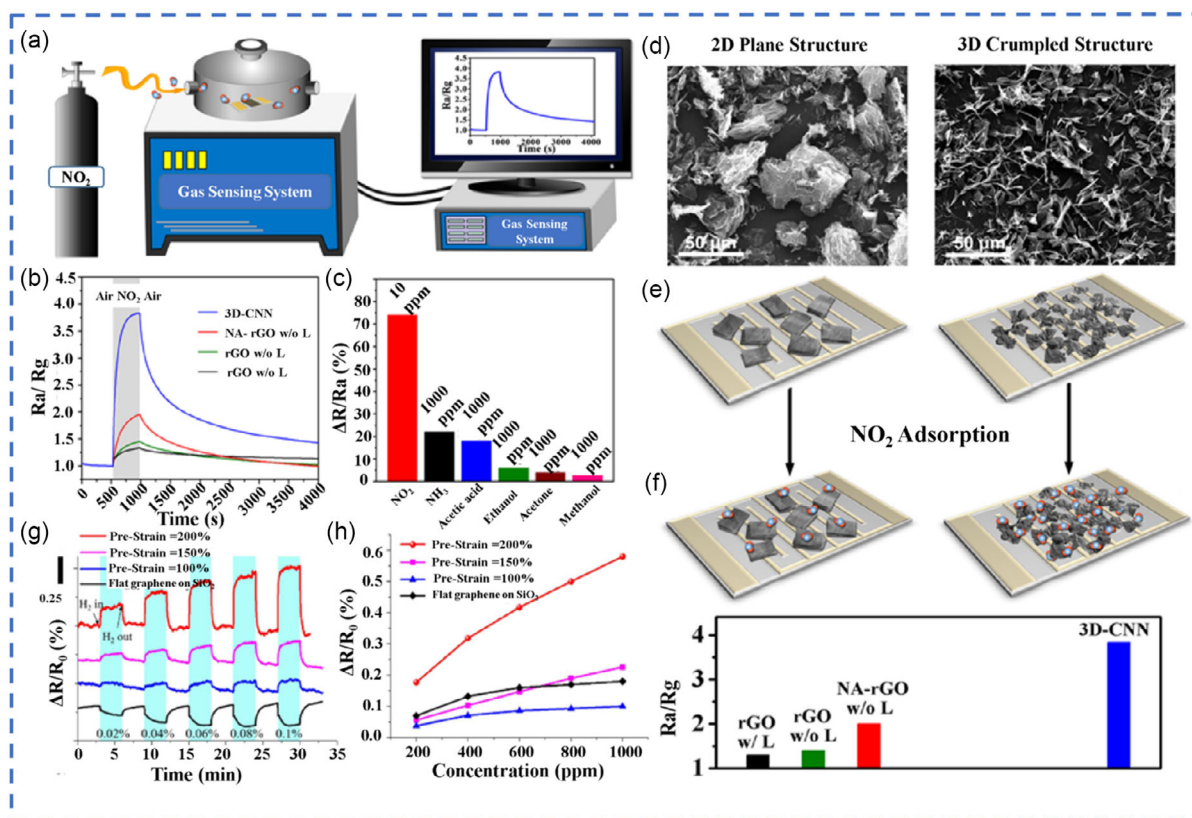


Figure 8. a) Flowchart of the optimal gas-sensing experiment. b) Comparison of the response of 3D-CNN with various rGO sensors to NO₂. c) Reaction selectivity of the 3D CNN sensors to NO₂ and various solvents. d) SEM images of 2D and 3D plane structures. e) Schematics with tightly aggregated and irregular crumpled nanosheets. f) Corresponding NO₂ molecule adsorption. Reproduced with permission.^[34] Copyright 2017, American Chemical Society. g) Hydrogen-sensing transient response curve. h) Sensitivity of crumpled and 2D graphene with varying prestrains on SiO₂ at ambient temperature. Reproduced with permission.^[82] Copyright 2019, IEEE.

exceptional selectiveness (Figure 8b) and strong reaction to NO₂ among the interfacial gases shown in Figure 8c, as calculated by the response $= \Delta R/R_a = (R_a - R_g)/R_a$. The study revealed that 3D CNN sensors are highly responsive to NO₂. The author stated that this is because sulfonate compounds have a high capacity for absorbing NO₂ molecules. The interfacial gases' poor charge carrier ability prevents a substantial shift in sensor resistance owing to NO₂. SEM image (Figure 8d) of rGO w/L shows tight aggregation blocks due to π - π interaction. In 3D CNN, nano-sheets are transformed from the 2D plane into the 3D structure by weird contortion and crumple right side (Figure 8e). Because of the extra space between crumpled sheets, more NO₂ molecules may be absorbed by the inner and outer surfaces (Figure 8f). Due to the production of multiple contortions and crumples, the plate distance between 3D CNN decreases, and the strength of the electric field changes at a faster rate along the whole, effective area, as seen in the provided diagram.^[34] Fei et al. found that NO₂ negatively affects sensor resistance, confirming graphene's p-type properties and NO₂'s electron-accepting role. When NO₂ gets absorbed into graphene, electrons are transported through the graphene to the NO₂ atoms, increasing hole concentration and decreasing resistance.^[139] However, the sensors' sensitivity is still a challenge for a researcher, as it has been found that sensitivity reduces as time progresses.

Furthermore, Shao et al. elucidate a flexible hydrogen gas sensor. The substantial curvature of CG particularly disrupts the graphene bonds and provides more significant contact with gas molecules, considerably raising one's sensitivity to hydrogen at ambient temperature. Hydrogen combines with oxygen ions that are firmly deposited on the CG surfaces owing to the disrupted π bonds and transfers electrons to the graphene. Their research shows the gas-sensing potential of CG devices in a hydrogen gas environment with biaxial prestrains ranging from 100% to 200% with flat graphene on SiO₂. Figure 8g shows that the resistance for flat graphene decreased when exposed to hydrogen and increased for CG. Figure 8h shows that the hydrogen sensitivity of CG with 200% prestrain increased by 320% compared with the flat graphene and 580% compared with CG with a 100% prestrain.^[82] According to Kothandam et al., the best adsorption energy is displayed by graphene flaws because of their larger active surface area.^[140] In addition, it is anticipated that the along with-deposited CGO thin films with the highest amount of functional groups that contain oxygen would have superior performance.^[53]

3.3. Strain Sensor

CG has many excellent properties, including ultrathinness, high elasticity, and ease of application to human skin. Strain sensors analyze strain fluctuations and detect local deformation and damage signals. Traditional sensors only work in one direction, are difficult to attach to flexible surfaces, and are not usually made to operate with living things.^[141–144] Fu et al. demonstrate a susceptible strain sensor using a single layer of graphene.^[145] Wang et al. showed that achieving a high strain of $\approx 30\%$ is possible using a completely reversible process.^[146]

Bai et al. described a new strategy for transferring and processing CG. They embedded graphene/PDMS in athletic tape attached to a human finger to monitor bending motions. Figure 9a,b shows that the sensor resistance increased with the bending motion of the finger. Each peak in Figure 9c represents a finger-bending movement and the degree of bending. Figure 9d shows the continuously bent and straightened finger results, which reveal that the CG strain sensor worked effectively for more than 200 cycles without any problems. The author's described method restricts the use of crumpled graphene in wearable electronics, such as human strain sensing, since it entails many phases of preparation, and strain sensor manufacture requires very stretchable substrates.^[147] The electrical response pattern of CG at a low strain limit may be attributed to the slide and stretching of ridges and crests, which have little impact on the change in resistance. As strains surpassed the typical impedance range of the sample, the resistivity of the CG increased more rapidly and irreversibly. The graphene structure is deteriorating irreversibly as a result of this nature. Consequently, controlling the maximum deformation within the longitudinal strained-resistance region is essential for the long-term application of CG in strain sensing. Therefore, the transfer procedure is crucial in applications involving crumpled graphene strain sensors.

Jin et al. experimented with a flexible strain sensor using CG. They showed that the applied tensile strain influences the resistance of graphene. They further developed a method to convert regular graphene into CG using a thin sheet of stretched silicone. They claimed that wrinkle formation was caused by an imbalance between the elasticity of graphene and the substrate.^[148] Because of their dependence on strain-induced control, the deformation mechanisms of materials linked to sensitivity still need to be explored.

Instinctive crumples/wrinkles created by compression and tension in layered systems may generate cheap and controlled

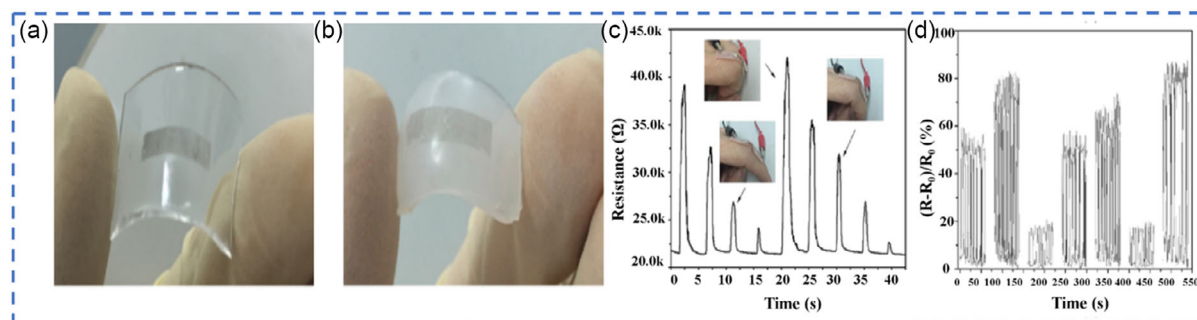


Figure 9. Spectral representations of graphene: a) PDMS and b) EcoFlex; c) the equivalent resistance varies in response to finger bends; d) the outcome of human motion monitoring reliability analysis. Reproduced with permission.^[147] Copyright 2017, Elsevier.

nanostructures for flexible sensors. Wrinkle patterns appear in a bilayer system owing to surface wrinkle instabilities; they must have managed to create ordered microstructures with tunable wavelengths and amplitudes since increased elasticity may lead to microcracks formation inside the material. Additionally, the necessity for controlling motion devices demonstrates that crumple/wrinkle graphene has significant promise in prosthetic limbs and intelligent robots, making this application of flexible sensors increasingly crucial in the future.

3.4. Chemical Sensors

Among the shortcomings of graphene synthesis procedures are multiple synthesis stages, a lengthy synthesis time, limited stability, stacking, and poor ease of processing for mass manufacturing.^[149–152]

Nonaka et al. investigated the problem of graphene restacking by synthesizing a CG structure using manganese ferrite NPs in a single step. They aimed to develop a new form of CG fully decorated with manganese ferrite NPs for H_2O_2 sensing. Spraying a precursor into the heating chamber at 400°C produces the CGB-like shapes seen in SEM micrographs (Figure 10a). Whereas in the morphology of the nanocomposites CG:Mn:Fe (1:5:5), cavities found in the bare CG sample were sealed with manganese ferrite, specifying that the CG surface was coated with metal oxide NPs (Figure 10b). In addition, TEM images of the CG in a 1:5:5 composite also indicate (dark spots) that it is covered

with MnFe_2O_4 NPs (Figure 10c). The GO and metal precursors in each aerosol droplet serve as a reactor, limiting the spread of the MnFe_2O_4 NPs over the CG. The author claims that this technique is a driving force in modifying the form of GO and regulating the particle size of metal oxides. The electrochemical behavior of graphene and CG/ MnFe_2O_4 composites was directly proportional to the quantity of MnFe_2O_4 on graphene. Figure 10d shows the interaction between CG and MnFe_2O_4 NPs, a strange composite phenomenon at a mean mixing ratio of 50 cyclic voltammograms. This later manifested as a subtle adjustment toward voltage regulators with the composite proportions 1:1:1 and 1:5:5. The experiment revealed that the strength at the composite ratio 1:1:1 (87%) was higher than that at 1:5:5 and the composite CG:Mn:Fe 1:1/2:1/2 also exhibited unusual behavior with an increment in current density along the cyclic voltammograms. The electrochemical stability of the composite 1:5:5 suddenly dropped after the first 10 cycles, attaining 61% after 50 cycles (Figure 10e). The lack of reduction in current density can be explained: by increased contact between both the carbon nanostructure and MnFe_2O_4 NPs as a consequence of the reduction of NPs that cover the exterior of the CG in the 1:1/2:1/2 composite. The increase in metal content in nanocomposites must necessarily reduce their specific surface area. Notably, the CG may capture analytical substances within its porous framework, thereby enhancing biomolecule–substrate electron transfer and electrochemical detection. However, the relationship between structure and electrochemical interaction should involve a complex interaction among surface area, pore sizes,

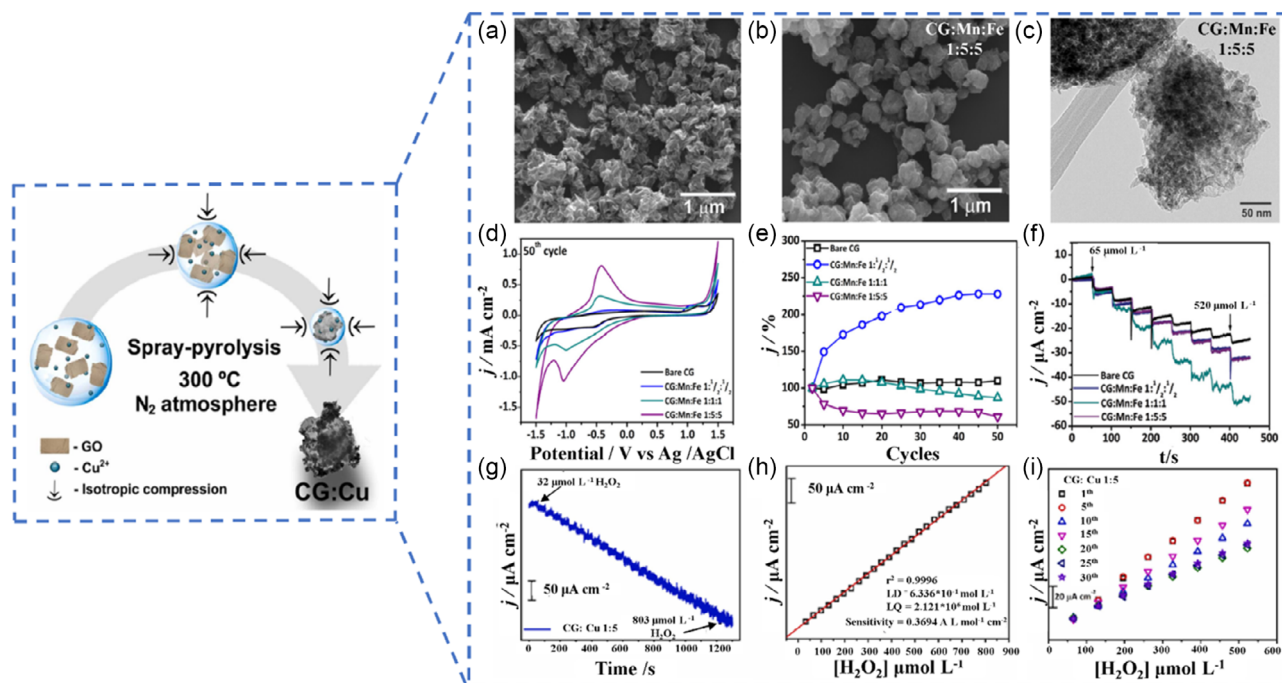


Figure 10. Illustration of the approach of crumpled graphene/copper-based nanocomposites. Reproduced with permission.^[154] Copyright 2022, Elsevier. SEM micrograph of a) bare CG; b) CG:Mn: Fe (1:5:5) composite; c) high-resolution transmission electron micrograph of CG:Mn: Fe (1:5:5); d) cyclic voltammograms (50th cycle); e) materials' electrochemical persistence after 50 cyclic voltammetric measurements; f) chronoamperometric response for H_2O_2 detection (from 65 to $520\ \mu\text{mol L}^{-1}$). Reproduced with permission.^[153] Copyright 2020, American Chemical Society. g) H_2O_2 intensity spectrum monitoring ($32\text{--}803\ \mu\text{mol L}^{-1}$); h) analytical curve generated based on (g) and i) H_2O_2 calibration curves gathered throughout 30 d. Reproduced with permission.^[154] Copyright 2022, Elsevier.^[154]

and graphene crystalline size arrangements, which needs further study.

The quasiaelectrochemical reaction to H_2O_2 is shown as an amperometric curve in Figure 10f. The composite 1:1:1 CG/metal ratio provides the best detection performance, whereas bare CG provides the worst. The synergy between the effective MnFe_2O_4 nanocrystals embellishing the conductive texture of crumpled graphene, resulting from a high number of readily accessible reaction sites facilitated by synthesizing of these crystals together with CG, is responsible for the best performance observed on nanocomposite 1:1:1.^[153]

Moreover, Alencar et al. reported a single-step copper-based NPs/CG 3D composite preparation. They explored the potential applications of the composites in H_2O_2 detection, in which low LD and excellent sensitivity parameters depended on the CG and copper (CG:Cu) ratio. They also determined the sensing ability of the composite sensor for 30 d. Figure 10g shows the composite CG:Cu (1:5) measurement at low H_2O_2 concentrations. In comparison, Figure 10h depicts an excellent linear bandwidth with a determination coefficient of ≈ 1 , with a smaller LD and sensitivity. The LD was determined using Equation (1), where s and S indicate each material's standard deviation and sensitivity. Surface aggregation, common for NPs' development in 2D graphene, is not seen with 3D crumpled graphene. This feature permits increasing the deposition of NPs on the surface without affecting the surface area, which is also observable with increased sensitivity. Cu-based NPs stimulate the electrochemical process while the ball-like 3D design could retain the targeted analyte. This "cage" effect raises the collisions, accelerates the reactions, and reduces the likelihood of substances passing through the 3D structure intact.

$$\text{LD} = 3s/S \quad (7)$$

They recorded H_2O_2 sensing data every 5 days a month (Figure 10i). In the initial 15 d, the modified electrode displayed high reproducibility and a tolerable lifespan for H_2O_2 detection. High H_2O_2 concentrations resulted in slight shifts in the curves. In addition to sensing, copper chloride hydroxide hydrate/CG composite allowed for its usage in heterogeneous photocatalysis and energy transmission.^[154]

Modified graphene's electrical conductivity is sensitive to external variables such as humidity, temperature, persistent charges build-up, and impurity, making accurate and repeatable sensing challenging. The method and its application still face challenges due to the high thermal energy needed to convert plain graphene to crumpled graphene, the need to functionalize graphene surfaces to regulate target molecule binding energy, and the need for an adaptable, responsive system that can be used across multiple fields. Table 1 shows a performance comparison of various CG sensors with materials required and methods involved for synthesis. A different application of crumpled graphene and its sensing ability show a lower LD from micrometer to nanosize. It is also observed that the crumple structure and material, including the fabrication process, influence the sensing performance. With increased lateral dimensions, plain graphene behaves like a fragile material with a self-folding or scrolling action. However, a nanoscale thin-layer wrinkled and

fragmented approach can accurately quantify the modulus of elasticity, durability, and cracking strain.

In terms of sensing performance, the performance of the biosensor and the gas sensor is quite remarkable. The curved shape of ridges facilitates the attachment of molecules or atoms and higher chemical reactivity. An essential parameter for the molecular activity of disorganized graphene is the feature of the number of ripples present on the surface. Graphene transfer wrinkles are seen to work as nanosized gas inlets to speed up chemical reactions under graphene using their 1D hollow and curved structures. The lattice's edges serve an essential function in chemical reactivity. To determine the chemical properties of defective systems, the crystallographic orientation of graphene is of the utmost importance. Although the strain sensor and the chemical sensor exhibit strong sensing abilities, as shown in the table, it is challenging to evaluate the performance of these sensors to that of other sensors due to the need for more data. For these applications, twisted and deformed graphene is beneficial because it makes the sheets more flexible while preventing layering and maintaining a greater surface area for a chemical reaction to occur.

3.5. Other Applications

H_2O_2 , an environmentally friendly oxidizing agent, is being produced more economically due to environmental concerns.^[155,156] H_2O_2 potentially operates fuel cells instead of hydrogen.^[157] Besides, the production of H_2O_2 is dependent on parameters such as infrastructure, chemical availability, and large-scale production. Researchers are continuously trying to develop new techniques to overcome these issues.

Lee et al. proposed a 3D construction to improve the active surface area and minimize mass transfer resistance for effective oxygen reduction reactions. Since all of the oxidized CG (OCG) had a wide D peak and acute G peak in the Raman spectra, we may extrapolate structural flaws in the system (Figure 11a). More defect sites are formed when oxygen functional groups are thermally reduced, which also result in the production of O_2 and CO_2 products. Figure 11b shows optimized oxygen functional groups and defect structures of OCG-800. In contrast, OCG-500 and OCG-1000 exhibited low H_2O_2 sensitivity, whereas OCG-250 had a minimal H_2O_2 sensitivity of 60–78%. OCG-800's superior electrochemical behavior may be explained by its 3D graphene surface, which has tailored oxygen functional groups and defect sites and facilitates mass transport of the reactant molecules to the active sites. They also performed density functional theory to predict the sensitivity of functionalized carbon catalyst and binding energy ΔG_{OOH}^* and $\Delta G_{\text{H}_2\text{O}_2}^* - \Delta G_{\text{O}}^*$, the active catalytic sites for H_2O_2 production. The exergy diagram (Figure 11d) further revealed that both CO and C–O–C had appropriate OOH adsorbents energy and resources for H_2O_2 generation, while C–O–C tends to progress to ORR because of its high oxygen adsorption. Figure 11c shows the active sites of the carbon atom right next to the oxygen in the ether group, with moderate OOH adsorption strength and potential H_2O_2 production.^[158] The author claims that OCG synthesized at 800 °C provides the ideal architecture for optimal loading circumstances but fails to recognize the mechanism to control defect production or the appropriate temperature for various metals catalysts.

Table 1. Comparison of CG graphene-based sensors performance.

Materials required	Methods	Limit of detection (LD) H/Response	Application	Merits of the seniors	References
Monolayer graphene, copper foil	CVD, annealing	$\approx 10\text{--}18 \text{ mol L}^{-1}$	FET Biosensor (nucleic acids detection)	As the electrical band gap shifts, rising molecular residence time due to deformed graphene leads to electrical hotspots, enabling attomolar detection of DNA and early identification of biomarkers associated with diverse human diseases.	[125]
Single layer graphene, PMMA, Acetic acid, RT-LAMP primer	Viral Transport Media (VTM)	44 nM	Biosensor (deAcetic sars-cov-2 virus)	The biosensor reduces detection time and enables the fabrication of miniaturized, cost-effective cgFET sensors to detect diverse pathogens.	[107]
Crumpled graphene, PDMS substrate, TiO ₂ substrate	COMSOL Multiphysics simulations	$-2650 \text{ nm RIU}^{-1}$ on PDMS substrate $-4810 \text{ nm RIU}^{-1}$ on TiO ₂ substrate	Plasmonic Biosensors	Solid biomolecule-graphene resonance interaction, made possible by the CG, makes RI sensors with a wide range of settings for antigen-antibody relationship dynamics and ultrasensitive condition biomarker detection.	[130]
Crumpled graphene, IL-6 protein, DNA, RNA	Molecular dynamics simulations	25 aM (Dopamine) 1 aM (S-protein)	FET biosensors (Dopamine, IL-6 and SARS-CoV-2 Proteins detection)	The possibility of adjusting the Debye layer helps identify size-independent biomarkers and allows rapid detection of S and N proteins.	[128]
Graphene, nucleic acid, COVID-19 samples	Molecular electromechanical system	$\approx 20 \text{ copies mL}^{-1}$	FET biosensors (SARS-CoV-2 detection)	A molecular electrical/mechanical mechanism enabled FETs to acquire viral nucleic acids selectively and rapidly transduce signals, resulting in the rapid and susceptible detection of SARS-CoV-2.	[108]
Crumpled graphene, elastomer substrate, NO ₂	Vacuum filtration method	13 ppb	Gas Sensors (NO ₂ detection)	Pre-strained crumpled QD film provides a highly stretchable NO ₂ gas sensor with potential real-time environmental and health surveillance applications.	[133]
rGO, N-rGO, Sodium hydroxide, ammonia solution, NO gas	Hydrothermal method	$R/R_0 = 0.012$ $R/R_0 = 1.7$	Gas sensor (NO detection)	As the capture of ions in the N-doped conduction spectrum increases, the NO sensing properties increase, and the N-rGO sensor displays superior sensing stability compared to the rGO sensor.	[136]
rGO, 3-Aminopropyltrimethylsilane, dimethyl methyl phosphonate (DMMP), Ferrous sulfate, and phenylene diamine	Microfabrication process, thermal treatment	$\Delta R/R_0 = 2.21\text{--}8.95$	Gas sensor (DMMP detection)	Design and synthesizing oppositely charged GO and rGO to make a susceptible, 12 layers of porous graphene (r(GO)/rGO) gas sensor to detect DMMP shows with rising vapour content 1–50 ppm response of the sensor achieved 2.21 to 8.95%.	[137]
3D-CNN, NO ₂ gas	Lyophilization	$\Delta R/R_a = 0.74$	Gas sensor (NO ₂ detection)	3D crumpled graphene nanosheets with noncovalent bonding modification and high response and selectivity NO ₂ gas sensor inspire new bionic sensor and chemicals trace detection platform.	[34]
Crumpled graphene, Ecoflex elastomer substrate	CVD	$\Delta R/R_0 = 0.02\%$	Gas sensor (H ₂ detection)	200% pre-strain elastomeric base crumpled graphene gas sensor shows 320% more sensitivity than a flattened graphene sensor.	[82]
Graphene, polydimethylsiloxane (PDMS) thin film	Uniaxially pre-stretched very-high-bond (VHB) film	Maximum strain 135% with a Gauge Factor of 337.8	Strain sensor	A pair of CG nanosheets with a flat graphene film encased between them for a highly stretchable design with an extensive working range shows a maximum tensile strain of 135%.	[143]

Table 1. Continued.

Materials required	Methods	Limit of detection (LD) H/Response	Application	Merits of the seniors	References
Crumpled graphene, PDMS	Thermal reduction process	Maximum strain 100% at gauge factor of 7.1	Nano cellulose Strain Sensors	The graphene-based high-strain sensors built on flexible nano paper has a gauge factor of 7.1 at 100% strain, over 10 times greater than stretchable Carbon nanotube and silver nanowire sensors.	[144]
Crumpled graphene, copper-based NPs, N ₂ gas	Spray pyrolysis	0.64 $\mu\text{mol L}^{-1}$	Electro-chemical sensor (H ₂ O ₂ detection)	Extended durability and an extremely sensitive H ₂ O ₂ detector using crumpled graphene as a substrate for nucleation and the growth of Cu-based NPs.	[154]
Crumpled Graphene, Manganese Ferrite NPs, Teflon stainless steel autoclave	Spray pyrolysis	0.61 $\mu\text{mol L}^{-1}$	Electro-chemical sensor (H ₂ O ₂ detection)	A single-step hydrothermal approach that generates a CG ball structure with a minimal LOD and an extra sensitive electrochemical sensor reveals an energy storage device of the future.	[153]
Graphene, AuNP	CVD, biaxially pre-strained high bond	Upto 200% strain	Photodetector	Crumpled hybrid structures with the mechanical ability to stretch up to 200% have 12 times the photo response of plain graphene photodetectors.	[159]

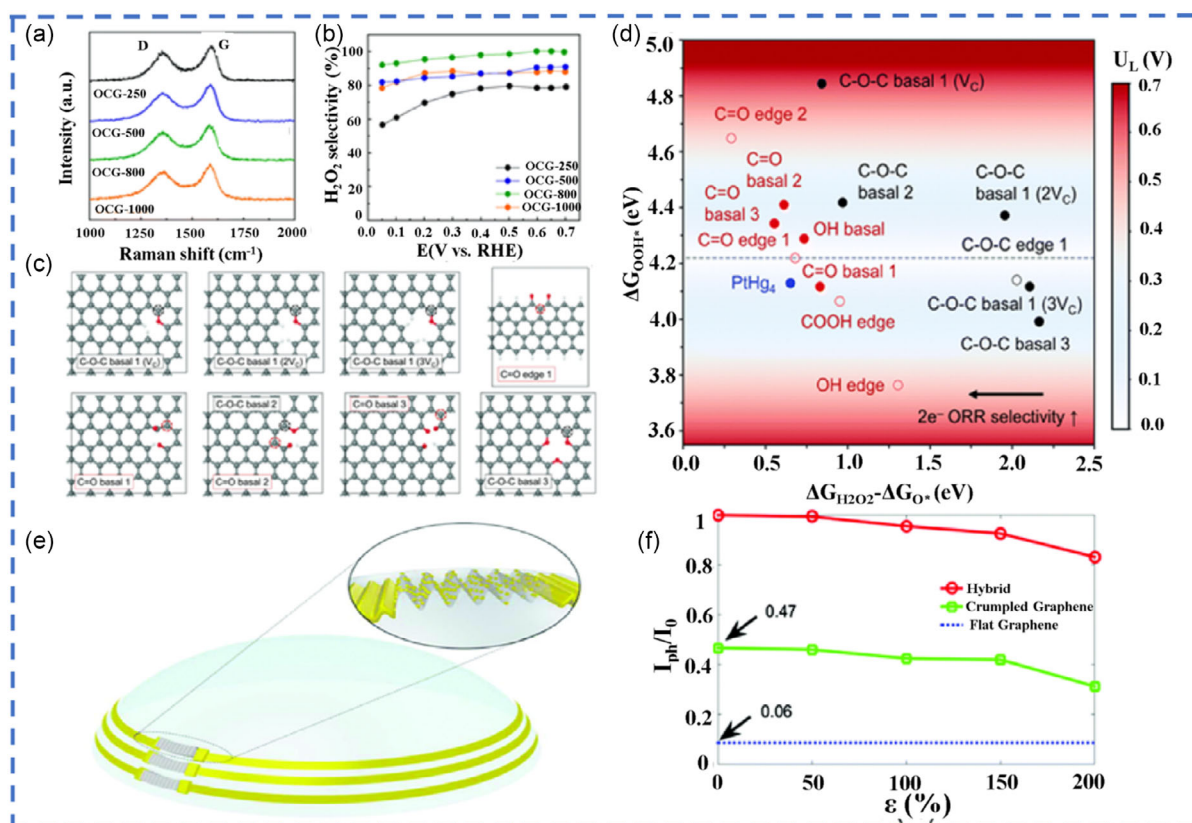


Figure 11. a) Raman spectra of oxidized crumpled graphene. b) H₂O₂ selectivity of OCG catalysts. c) Model system of various oxygen functional groups. d) Relationship of scale between ΔG_{OOH}^* and $\Delta G_{\text{H}_2\text{O}_2}^* - \Delta G_{\text{O}}^*$ of different oxygen functional groups. Reproduced with permission.^[156] Copyright 2022, Royal Society of Chemistry. e) Schematic represents a stretchable hybrid contact lens photodetector. f) Comparative representation of the standardized incident photons (I_{ph}/I_0) of a crumpled hybrid photodetector and 2D graphene devices at varying uniaxial tensile strain. Reproduced with permission.^[159] Copyright 2017, Royal Society of Chemistry.

Besides, Kim et al. developed a photodetector consisting of CG integrated with Au NPs, which enhances mechanical stretchability and photoresponsivity. Figure 11e demonstrates the photocatalytic activity characteristic of a flexible hybrid photosensor; the photocurrents measured by the crumpled hybrid photodiode were 1570% and 110%. They observed ≈ 11.99 times more photo-sensitivity of the crumpled photodetector than the CG photodetectors, as shown in Figure 11f.^[159] Researchers have noticed that crumpled graphene can be used as packaging for metal nanoclusters,^[158] where heteroatom doping also creates amorphous intrinsic defects. Suitable absorbent materials can significantly improve device rates, performance, and stability.^[160]

4. Future Scope of Crumpled Graphene

For the next decade, or until each of graphene's numerous potential uses meets its requirements, the market for graphene applications will be driven by progress in graphene production. A thermochemical-shock procedure is used to simultaneously do both exfoliation and reduction in economically significant production of GO and its derivatives. Even though the resultant substance might have deformed graphene components with multiple layers, it retains many desirable properties of graphene with a single layer. Although this review focused on various graphene derivatives and applications, low-cost graphene production in mass substantially impacts affordable crumpled graphene development and application. Moreover, uncertainty remains about producing the increasingly intricate structure and geometry for integrated sensors. Particularly, crumple generation and surface wrinkling continue to be challenging to characterize pattern creation at solid substrates. Moreover, crumple formation in graphene materials must be more precisely understood and fully controlled. The expert's opinion for the instability of a graphene atomic layer is due to environmental variations and random film flaws. Therefore, it is important to understand the approaches and strategies to control the crumpling of nanometer-to-subnanometer thin films. Crumpling for the strain engineering application of graphene materials is another promising approach. Nanobiotechnology, optoelectronic devices, and sensor manufacturing may benefit from dynamic surface crumpling with customizable functional features. While to increase the reversibility and responsiveness of devices, new approaches must be considered. Synthesis methods like laser ablation, thermally induced phase separation, and molecular adsorption could produce uniformly wrinkled structures.

Graphene 2D materials, depending on their form, exhibit different behaviors, such as the semiconductive and metallic (e.g., graphene) modification of energy bandgaps of graphene materials through surface wrinkling and crumpling formation. Consequently, various electromechanical properties of 2D materials could be accurately adjusted for sensor development. Rapid CG processing and production advances with customizable features will make effective multimodal sensing devices. However, more fabulously, imprinted graphene formations must be controlled for practical applications. Graphene materials put over well-designed structural and prestretched polymer substrates will permit controllable crumple structures. Because of distinctive atomic thicknesses, high conductivity, and energy band

configuration, various 2D substances besides activated carbon nanomaterials are being studied by researchers for their optoelectrical features. Hence, crumpling 2D materials gives excellent opportunities to expand their use in future advanced sensor systems.

5. Conclusion

Overall, this review provides insights into CG's material advancements and implementations in manufacturing various sensors. In addition, it explains the physical and mechanical features of different engineered crumpled surfaces to improve the sensitivity of multiple sensors. Despite the recent challenges and limitations in fabrication, introducing different surface-engineered materials and producing high-performance sensors by combining various forms of graphene and revolutionary micro/nano-technology amplify their capability in electrochemical sensors, gas sensors, and biosensors.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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