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Review Article

Graphene-Based Films as Effective Mosquito Repellent for Human Skin Protection: A Brief Review

Anuradha Parixit Prajapati[1,*], Shivani Gandhi[1], Garima Singh[1], Sachin Narkhede[2], Shailesh Luhar[3]

[1]Department of Pharmaceutics, Smt. BNB Swaminarayan Pharmacy College, Salvav, Vapi, Gujarat, INDIA.
[2]Department of Pharmacognosy, Smt. BNB Swaminarayan Pharmacy College, Salvav, Vapi, Gujarat, INDIA.
[3]Department of Quality Assurance, Smt. BNB Swaminarayan Pharmacy College, Salvav, Vapi, Gujarat, INDIA.

ABSTRACT

Mosquito-borne diseases pose significant public health threats worldwide, necessitating effective prevention strategies. Traditional chemical agents used for mosquito bite prevention often carry environmental and health risks. In this review, we explore the potential of graphene-based films as a non-chemical approach for protecting against mosquito bites. The aim of this review is to assess the effectiveness of multilayer graphene films in preventing mosquito bites and to explore their practical implications for public health. Through experimental investigations, researchers found that dry multilayer graphene films effectively block mosquitoes' ability to detect skin or sweat chemicals, thereby preventing mosquito bites. Additionally, these films can serve as physical barriers to the mosquito's feeding mechanism. The findings suggest promising applications of graphene films in protective technologies for human skin and smart fabrics. Graphene's non-toxic nature and ease of application make it an attractive alternative to chemical repellents. Implementing graphene-based films for mosquito bite prevention could potentially reduce the transmission of mosquito-borne diseases, addressing critical public health concerns. In conclusion, this review highlights the potential of graphene films as a non-chemical method for mosquito bite prevention. Future research should focus on evaluating the long-term effectiveness and safety of graphene films, paving the way for the development of innovative technologies that utilize graphene to safeguard against mosquito bites and mitigate the spread of infectious diseases.

Keywords: Graphene, Mosquito repellent, Chemo-sensing, Molecular barrier, Puncture resistant' s.

Correspondence:
Dr. Anuradha Parixit Prajapati
Department of Pharmaceutics, Smt. BNB Swaminarayan Pharmacy College, Salvav, Vapi-396191, Gujarat, INDIA.
Email: anupatel03@gmail.com

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INTRODUCTION

Graphene, a two-dimensional material composed of hexagonally arranged carbon atoms, is a fundamental component of graphite and finds numerous applications in diverse fields such as batteries, pencils, and pharmaceuticals. This "wonder material" boasts exceptional properties that make it stand out among other materials.[1] For instance; it is incredibly thin, being just one carbon atom thick, yet remarkably strong, outclassing diamond and steel of similar thickness. Moreover, graphene is flexible, transparent, and possesses enormous surface area, making it an exceptionally stretchable crystal. Additionally, it is an excellent conductor of both electricity and heat, and exhibits fascinating optical properties.[2]

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The potential applications of graphene span across several industries, including electronics, optoelectronics, electrochemical batteries, and composites, among others. One particularly promising application of graphene is in the form of films, which have gained significant attention for practical uses, such as heat spreading in mobile phones. This has resulted in a growing interest in commercializing this technology, highlighting the immense potential of graphene for industrial and consumer applications.[3-8]

Use graphene, and while the industry is still growing, applications are starting to emerge.[9]

This review provides a comprehensive overview of the latest developments in the field of graphene-based film, covering various aspects such as fabrication, processing techniques, interfacial modification, and applications. The primary emphasis of this review is on the fabrication methods of graphene-based film, different approaches to interfacial modification, and techniques to enhance its mechanical properties. Additionally, the article also discusses the current challenges and status of applications of graphene-based film in energy storage, environmental, and smart-device applications. By summarizing the recent advancements, the review aims to provide valuable insights to researchers and scientists in the field of material science and engineering.[10]

DIFFERENT GRAPHENE MATERIALS

Monolayer graphene sheets

High-quality materials of mono layer graphene are produced through vacuum processes such as Chemical Vapor Deposition (CVD), wherein gaseous reactants are deposited onto a substrate to create a layer of graphene[11] in Figure 1.

Bilayer graphene

When two sheets of graphene are stacked on top of each other, they typically exhibit a zero bandgap. However, it is possible to introduce a controllable bandgap by applying an electric displacement field to the two layers. Another way to create a bandgap is by arranging the two layers in a specific manner during the stacking process.[12] This can be seen in Figure 2.

Few-layer graphene

Few-layer graphene refers to graphene materials that consist of three or more layers of graphene sheets stacked on top of each other. Few-layer graphene has unique properties that are distinct from those of single-layer graphene, such as increased mechanical strength and improved electrical conductivity. These properties make few-layer graphene a promising material for various applications, including electronic devices, energy storage, and sensors,[13] mostly observe in Figure 3.

Graphene Nanoribbons

(GNRs): Graphene nanoribbons (GNRs) are extremely thin (less than 50 nm) graphene strips. These depend on the material's width and edge type and have intriguing electronic properties (zigzag type or armchair type). GNRs are semiconductors with an inverse relationship between their energy gap and ribbon width.[14] This type can be seen in Figure 4.

Graphene Flakes/Nano Platelets (GNFs/GNPs)

Some of the mechanical, thermal, and electrical properties of graphene can be preserved in Graphene Flakes (GNFs) observed in Figure 5. GNFs can be synthesized in a variety of shapes and sizes. GNFs, a form of graphene that is relatively inexpensive, are being used more frequently in a variety of composite materials.[15]

Graphene Oxide (GO)

A three-dimensional substance called graphite is composed of many graphene layers. Graphite (carbon), hydrogen, and oxygen make up the chemical compound known as graphite oxide. The oxygen molecules in graphite oxide serve to separate the carbon layers (the graphene sheets) Figure 6. Graphene Oxide (GO), which is composed of single carbon, oxygen, and hydrogen sheets, is produced when graphite oxide is dissolved in water. Other than grape, graphene oxide can be used for applications due to its unique characteristics.[16]

Reduced Graphene Oxide

Reduced Graphene Oxide Graphene oxide sheets can be reduced (which means removing the oxygen and hydrogen) to get regular graphene sheets.[17]

Preparation Techniques of Graphene-Based Film

Graphene-based films can be prepared using several methods including vacuum filtration, evaporation-induced self-assembly, rod coating, spin coating, and electro-induced preparation.

Vacuum Filtration

Guo *et al*. have developed a new method to create composite films with insulating properties using vacuum filtration. While graphene-based films have shown promise in various applications, their high electrical conductivity can limit their use in certain settings. To address this issue, the researchers added Al_2O_3 to the cellulose nanofiber/graphene-based composite film during the vacuum filtration process. This addition not only improved the insulating properties of the film, but also increased its thermal transfer capabilities. Furthermore, the team

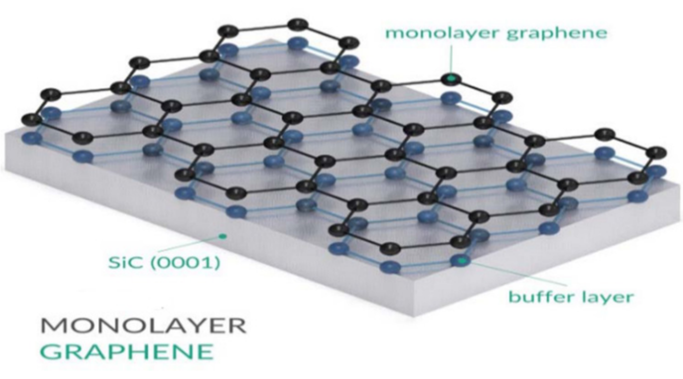

Figure 1: Monolayer sheet.[11]

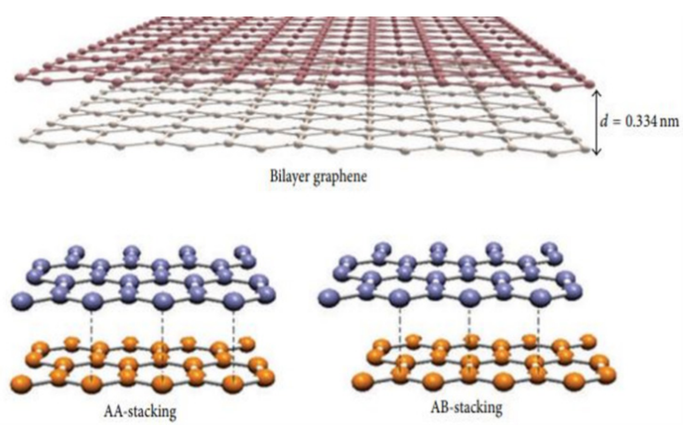

Figure 2: Bilayer graphene.[12]

employed a high-temperature annealing approach to carbonize polydopamine nanoparticles, resulting in a graphene film with exceptional thermal conductivity observe in Figure 7. These innovations have the potential to enable new applications for graphene-based films in a variety of industries.[18-20]

Vacuum filtration is a commonly used technique in the laboratory for separating solid particles from a liquid solution, which can produce films of varying thicknesses by optimizing the concentration. However, this method has limitations when it comes to industrial-scale production. As the thickness of the film increases, the filtration efficiency decreases due to the clogging of the filter, which can make it challenging to produce thick films within a reasonable time. Additionally, the size of the film is restricted by the filter's size, making it less suitable for large-scale production. As a result, alternative filtration techniques such as centrifugal, membrane, or continuous filtration may be more appropriate for industrial-scale production, as they offer higher throughput and more consistent results. It's essential to consider the specific filtration requirements and choose the most appropriate technique accordingly.

Evaporation-Induced Self-Assembly

By pouring a slurry into a polytetrafluoroethylene mould and allowing the solvent to evaporate at either ambient temperature or a specified temperature, flexible free-standing macroscopic graphene-based sheets are produced.[21] After that, the substrate is pulled away from the resultant film. The capillary-forced-assisted self-assembly approach, which starts the film at the contact line of the air-liquid-solid interface, is another technique for creating reduced graphene oxide films with unidirectional organization. Large-scale highly organized films are created using this approach.[22]

By altering the concentration and volume of the GO suspension, researchers have also created Graphene Films (GFs) on an aluminium substrate via an evaporation self-assembly technique.[22] The produced GFs surpass Polylitic Graphite Sheets (PGS) by 60% and have a high thermal conductivity of 3200 W m1 K1 with a thickness as thin as 0.8 m and which outperformed the Polylitic Graphite Sheets (PGS) by 60%. Figure 8, GFs have great mechanical tensile strength and outstanding flexibility.

The use of evaporation-induced self-assembly in the fabrication of graphene-based films has enormous promise. This method is simple to use and doesn't need any special tools. The substrate's size can be changed to produce films of various thicknesses.[23] The films created using this process also have smoother top surfaces than those made using vacuum filtering. However, it's crucial to be aware that bubbles could occur during the evaporation process and damage the film's structure.

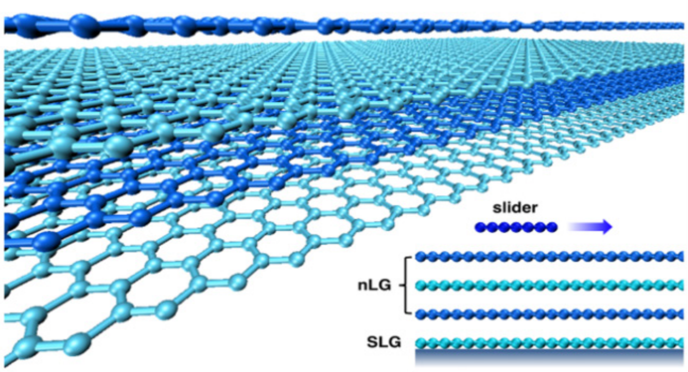

Figure 3: Few layer graphene.[13]

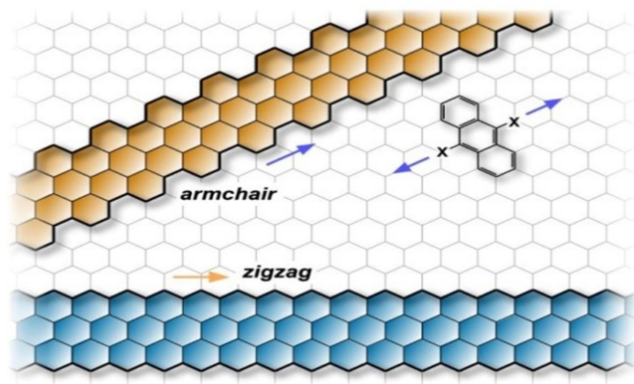

Figure 4: Graphene Nanoribbons.[14]

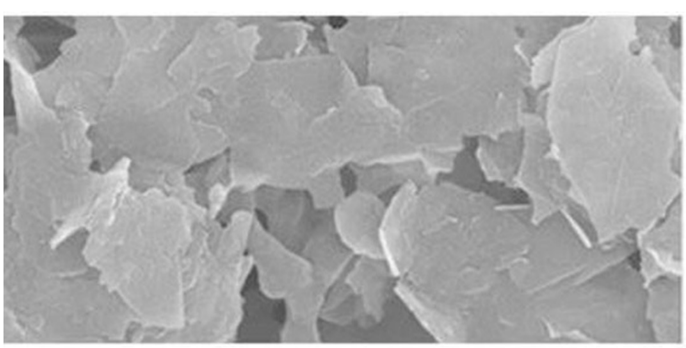

Figure 5: Graphene flakes.[15]

Figure 6: Graphene Oxide.[16]

Spin Coating

A common approach for producing sheets made of graphene is spin coating.[21] With this technique, a solution is deposited onto a substrate, and centrifugal force is used to create a thin film. Spin coating is a well-liked technique for Layer-by-Layer construction, which may quickly deposit a highly ordered multilayer internal structure.[24] GO nanosheets have a weak van der Waals connection, which can cause stacked GO layers to easily split during the thermal reduction process. This causes many voids and lowers the films' cross-plane thermal conductivity.[24,25]

Spin-Assist LBL (SA-LBL) has been created to address this problem. To create robust internal bonding multilayers, this technique alternates electrostatic deposition between complementary charged elements. For instance, Hong *et al.*

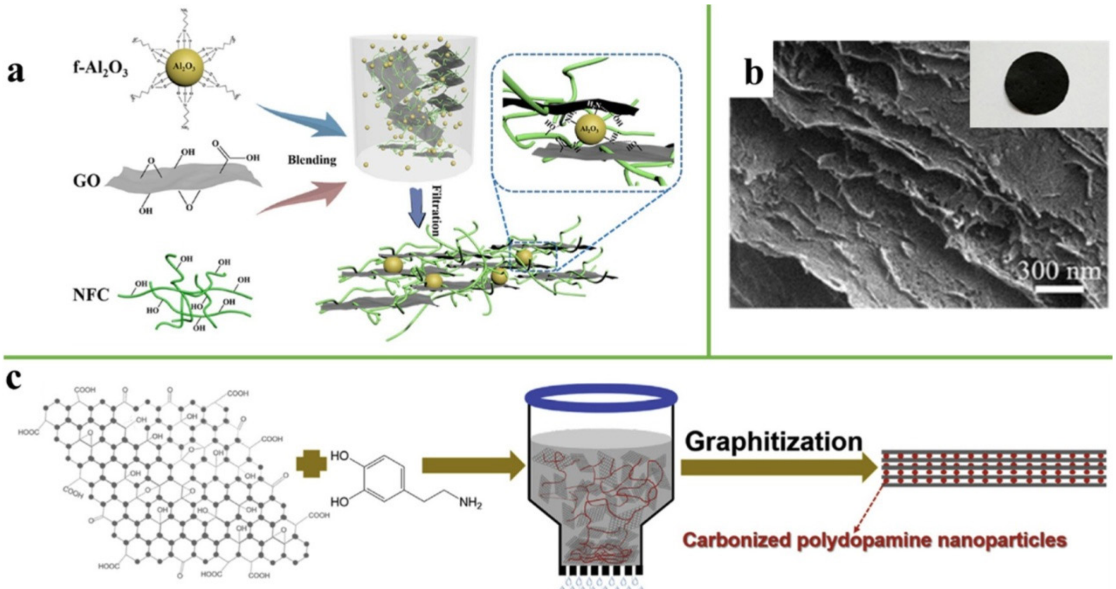

Figure 7: (a) Schematic illustration of the fabrication process of a f-Al_2O_3@RGO/NFC composite film by vacuum filtration.[16] (b) Macroscopic photograph and cross-sectional SEM views of composite film. (c)Schematic illustration of GF.[16]

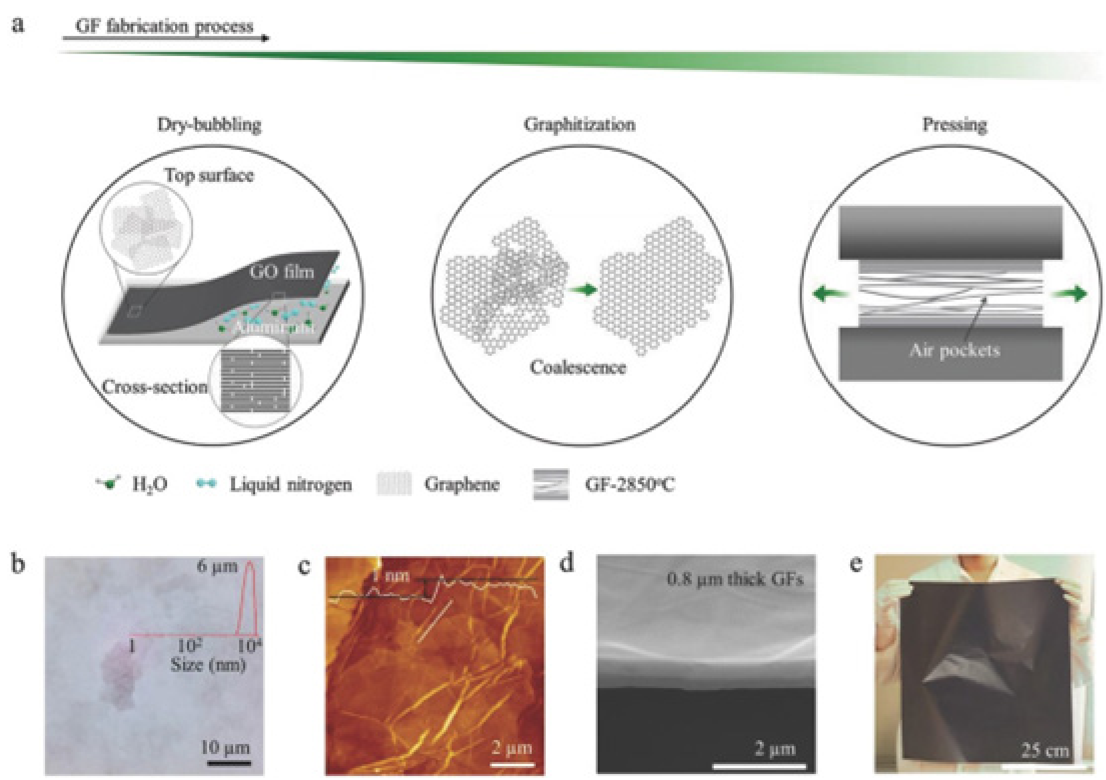

Figure 8: (a) Sketch of the fabrication process of GF. (b) Optical image of GO flakes with an average size of about 6 µm. (c) AFM image of GO flakes with a thickness of less than 1 nm.[20]

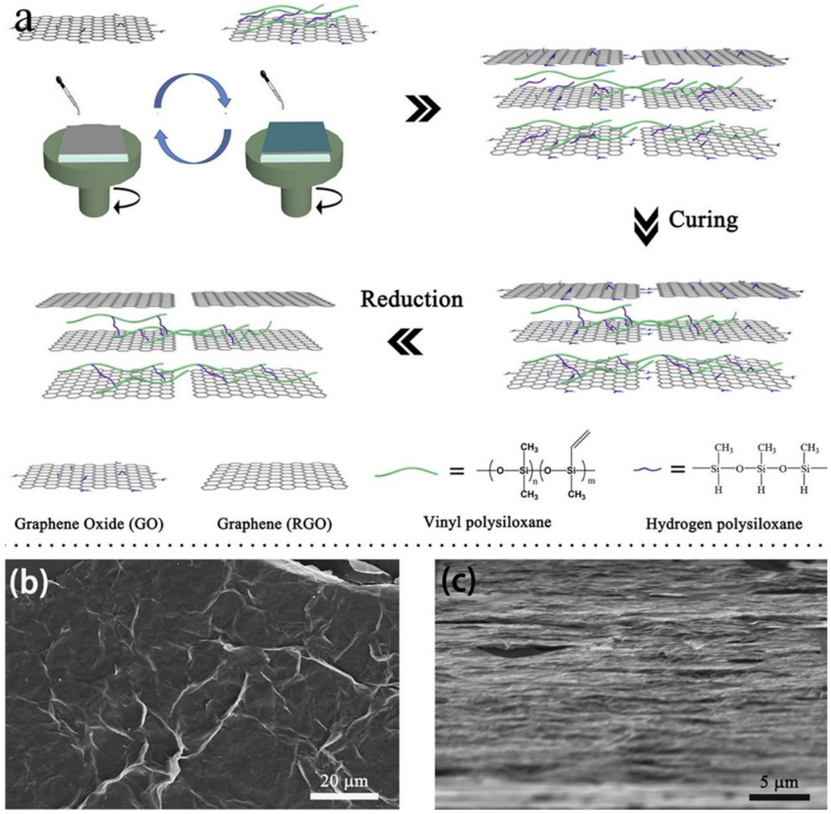

Figure 9: (a) Schematic depiction of the suggested spin-assisted LBL assembly process for the creation of multilayered SR/RGO films. GO on SR as seen in (b) via SEM. (c) SEM picture of the SR/RGO 40 film in cross-section.[25]

improved the in-plane and out-of-plane thermal conductivity of the composite film by fabricating a rGO/alumina film using SA-LBL.[24] Song *et al.* employed the SA-LBL procedure to make the silicone rubber/graphene film depicted in Figure 9 to show that the SA-LBL procedure may also produce hydrogen bonding contacts. The composite film's thermal conductivity improved as a result of the combined hydrogen bonding and van der Waals forces that increased bridging between SR and GO.[25]

Spin coating is a widely used technique for creating ordered multi-layered films because it is straightforward, versatile, and inexpensive, although it should be noted that spin coating is less effective than blade coating. Spin coating can only apply a very thin film at a time, therefore many applications are necessary to reach the appropriate thickness. Because of this, it is inappropriate for large-scale industrial production when speed and effectiveness are important considerations. Blade coating is a more effective technique for creating films of a particular thickness quickly than spin coating.[26]

Blade Coating

Blade coating is a common technique used in both laboratory and industrial environments to deposit a slurry onto a substrate. In laboratory settings, this process involves moving a blade over the substrate to transfer the slurry onto it. However, in industrial applications, the slurry is typically transferred to a fabric substrate that is then processed through multiple stages of heat treatment to create a freestanding film roll. By adjusting the interval between the scraping blade and the substrate, the thickness of the resulting film can be controlled.[27]

Chen *et al.* conducted a study where they utilized blade coating process to produce large-sized ultra-thick and dense laminated-structures Graphene Films (GFs) in a quasi-industrial scale. These GFs were then incorporated into a standard mobile phone template and exposed to a constant heat source. The GFs displayed faster thermal transfer performance when compared to PGF. Additionally, the GFs' surface maintained their original structure even after undergoing a bending test, which did not negatively impact the heat diffusion process. This result was illustrated in Figure 10 of the study.

Blade coating is indeed a popular and efficient method for producing graphene films, as you mentioned. However, it does come with certain challenges, such as the strict requirements for slurry concentration and viscosity.[27]

To elaborate further, blade coating involves spreading a liquid mixture (slurry) containing graphene particles onto a substrate using a blade. The thickness of the resulting film is determined by the speed and pressure of the blade, as well as the viscosity of the slurry. If the viscosity is too low, the slurry will not spread evenly, resulting in patchy or incomplete coating. Conversely, if the

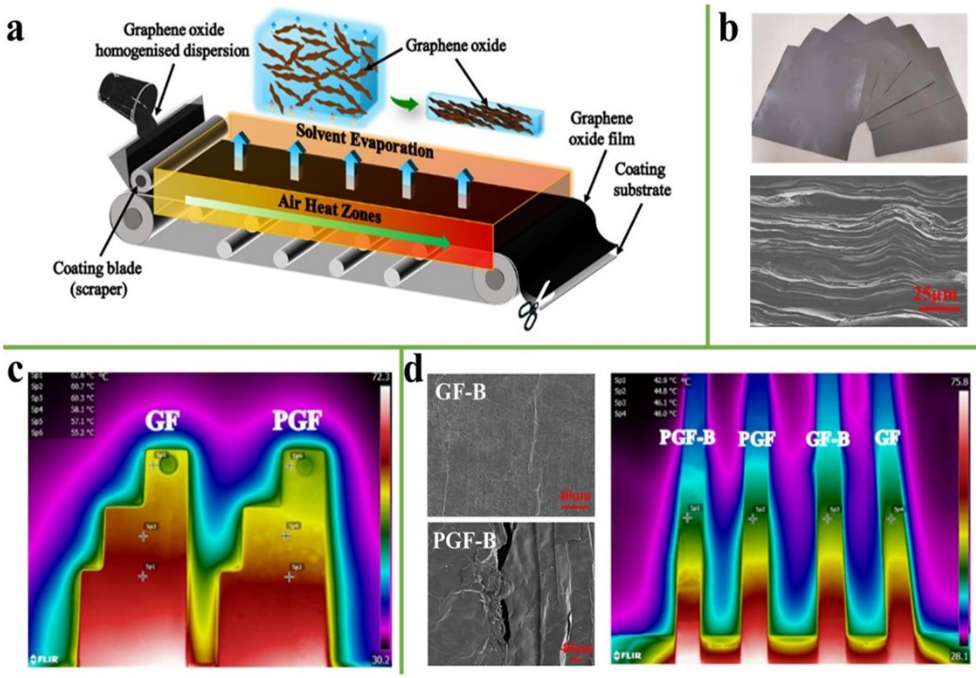

Figure 10: (a) The Schematic of the fabrication process of the GO film. (b) Optical image of the GFs with different thicknesses and cross-sectional SEM image of pressed GFs-2850. (c) Infrared thermal images of 220 μm GF and PGF in a standard template. (d) SEM image of the surface morphology change of GF and PGF after the bending test, respectively, and infrared thermal images of GF and PGF before and after the bending test.[27]

viscosity is too high, the slurry will not flow smoothly, resulting in an uneven coating thickness.

To overcome these challenges, it is important to carefully control the properties of the slurry, including the concentration, viscosity, and surface tension. Additionally, the properties of the substrate and the blade must also be optimized to ensure uniform coating. With proper optimization and control, blade coating can produce high-quality graphene films with precise thickness and size control, making it an attractive option for industrial mass production.[28]Top of Form

Electro-Induced Preparation

For the purpose of producing graphene-based films, researchers are investigating the electro-deposition procedure. It involves causing the migration of charged particles in a stable suspension by applying an electric field between two electrodes.[28] Different foils can be produced by varying variables like current density, electro-deposition duration, and electrolyte content. Because it possesses a negative charge and strong hydrophilicity in water due to the presence of oxygen-containing functional groups, Graphene Oxide (GO) is often utilized as the precursor during this procedure see in Figure 11.

The presence of Cu atoms aids in the reduction of GO, and some of the oxygen-containing functional groups on GO can be removed via electrolysis of a Cu ion on the Cu anode.[29] This causes residual oxygen on Reduced Graphene Oxide (RGO) and Cu to form Cu-O-C bonds, improving the interfacial bonding strength of the Cu-RGO composites. Cu-RGO films have been successfully created by researchers utilizing GO suspension and DC voltage. These films have exceptional flexibility and heat-transfer capabilities. As thermal conduction routes are created in the film throughout the course of the deposition process, the Cu-RGO film's thermal conductivity rises.[30,31]

With any stable solution, electro-induced deposition is a flexible technology that has several advantages, such as accurate thickness control, good sample homogeneity, and a simple, economical operation method. However, it is crucial to use top-notch precursor solutions when using this approach to create graphene-based films. Otherwise, there is a chance of having problems with agglomeration or lower deposition efficiency.

Potential Applications

Graphene is a very versatile material that may be mixed with other substances to create a variety of new compounds with improved qualities. Researchers from all over the world are continually studying and patenting graphene to learn more about its many features and potential uses[33], which include:

- Batteries,
- Solar cell,
- Water filters,
- Super capacitors,
- Energy generation,

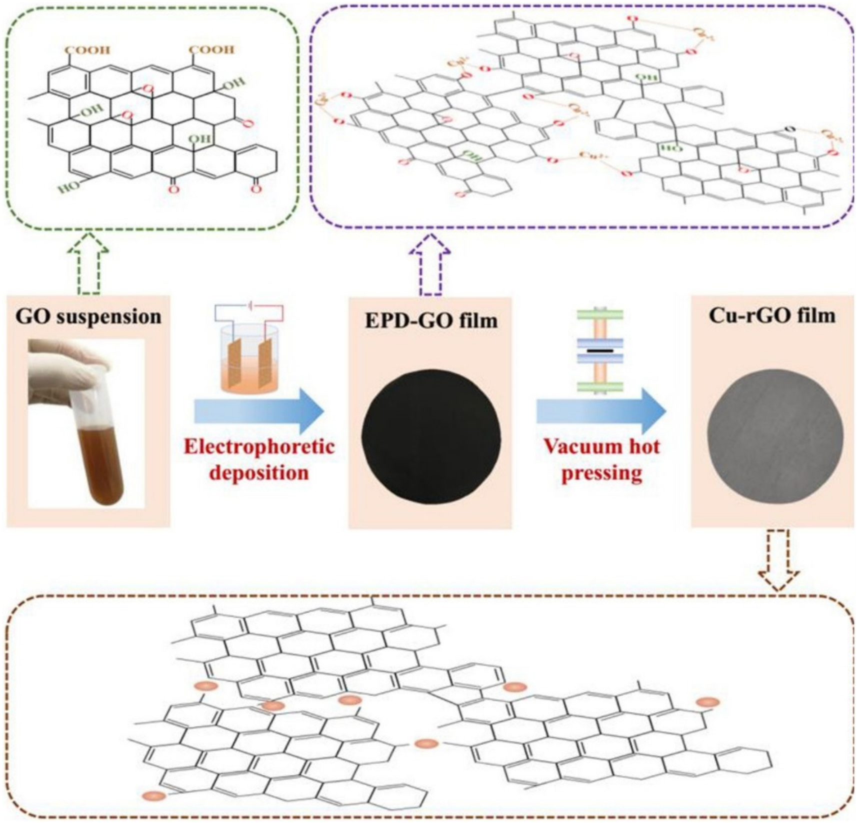

Figure 11: Schematic depicts the preparation process of composite film.[32]

- Lubricant,
- DNA sequencing,
- Mosquito repellent,
- Coating,
- Spintronics,
- Transistors,
- Computer chip,
- Drug delivery,
- Touch screen,
- Antennas.

The need for efficient electrochemical energy storage devices has become increasingly urgent as energy consumption continues to rise, and renewable energy development has become a crucial topic.[34] Graphene-based films are a promising material for super capacitors due to their high electrical conductivity, large specific surface area, and strong mechanical properties.[35,36] They have been shown to enhance energy storage capabilities and exhibit excellent power density and energy density. In addition, graphene-based films have also been used in flexible lithium-ion batteries as suitable electrode materials, which have good electron/ion conductivity, mechanical properties, and specific capacity. Graphene-based films have also gained attention in environmental applications, particularly in gas and water treatment, due to their unique layered and porous structure. They have demonstrated high hydrogen selectivity and water desalination performance. Moreover, graphene-based materials have been widely used in sensing applications, including chemical, bio-, and gas sensors, due to their high specific area, electron mobility, and low electrical noise.

Graphene-based films[37] have a wide range of potential applications due to their unique properties, including high mechanical strength, excellent electrical conductivity, and high surface area. Here are some examples of their applications:

Flexible graphene-based composite films[38] for Super capacitors: Graphene-based composite films can be used as super capacitor electrodes, which have high energy storage capacity and long cycle life. These films can be flexible and have high surface area, making them suitable for use in wearable electronics and other portable devices.

A free-standing sandwich-type GN/NC/Si Laminar Anode for flexible rechargeable Lithium-ion batteries: Graphene-based films can be used as anodes in lithium-ion batteries, which can be made flexible and lightweight by using graphene-based composite films. These films can improve the battery's performance by increasing its energy density, reducing its weight, and improving its stability.

High-performance Graphene Oxide[39] (GO) nanocomposite membrane for hydrogen separation: Graphene-based films can be used to create high-performance membranes for gas separation applications. The films have high permeability, selectivity, and mechanical strength, making them ideal for use in hydrogen separation applications.

Multilayer nonporous graphene membranes for water desalination:[40] Graphene-based films can be used to create membranes for water desalination applications. These films have high permeability and selectivity, making them ideal for removing salt and other impurities from seawater.

Novel graphene-based biosensor for early detection of Zika virus infection: Graphene-based films can be used to create biosensors that can detect viruses and other pathogens in biological samples. These films have high sensitivity and selectivity, making them ideal for use in medical diagnostics.

Graphene based electroluminescent gas[41] sensor for CO_2 detection at room temperature: Graphene-based films can be used to create gas sensors that can detect carbon dioxide and other gases at room temperature. These sensors have high sensitivity, selectivity, and stability, making them ideal for use in environmental monitoring and other applications.

Mosquito Repellent Action

The ultra-thin yet sturdy material acts as a physical barrier that mosquitoes cannot penetrate.

Graphene can provide a two-fold defence against mosquito bites acting as a mosquito repellent.

Graphene could provide protection from mosquitoes and help stop the spread of fatal mosquito-borne diseases like dengue, malaria, and chikungunya.

The chemical signals that mosquitoes use to detect the presence of a blood meal are blocked by graphene, which reduces their initial desire to bite.

According to researchers who have been working on fabrics that contain graphene as a barrier against hazardous chemicals, mosquitoes are a significant global disease vector and "there's a lot of interest in non-chemical mosquito bite protection."

Researchers discovered that mosquitoes could penetrate water-soaked graphene oxide coatings. However, they found that graphene with reduced oxygen content (rGO), when utilised as a biting barrier, functioned well in both dry and moist environments.[42]

CONCLUSION

The overview of graphene-based materials includes a wide range of topics, including numerous graphene materials, prospective uses, ways to make graphene film, and how it works to keep mosquitoes away. The review's conclusion highlights the following important ideas. A variety of graphene compounds, including graphene oxide, reduced graphene oxide, and graphene nanocomposites, are examined in the review. These materials may be customized for certain uses and have special qualities.

Examples of potential applications Numerous possible uses for graphene-based materials exist, including electronics, energy storage, sensors, and composites. Future technologies may benefit from their remarkable conductivity, mechanical strength, and flexibility.

The study examines several techniques for producing graphene sheets, such as chemical reduction, liquid-phase exfoliation, and Chemical Vapor Deposition (CVD). Each approach has benefits and drawbacks.

Graphene-based materials have been investigated in certain research as potential mosquito repellents. Even if the results are encouraging, more study is required to improve formulations and comprehend the mechanism of repellent activity.

In conclusion, the use of graphene-based materials as mosquito repellents is an intriguing field of study that has a lot of potential. To fully utilize their strengths in these several domains, additional research and development are needed.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interest.

ABBREVIATIONS

GNRs: Graphene nano ribbons; **GF:** Winged-helix; **CVD:** Vapor deposition; **GNFs:** Graphene flakes; **GO:** Graphene oxide; **rGO:** Reduced oxygen content.

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