

A review of microstructure of Graphene
And its future applications

Abstract

Scientists have tried to examine graphene's applicability in several sectors in recent years, due to its great features, and numerous literatures are well published. The fundamental unit is the same for simple two-dimensional graphene to complicated three-dimensional graphene linked structures, but they can have varied morphologies, microstructures, and characteristics due to microstructural design and play distinct roles in different applications. The current research in graphene microstructure engineering for improving performance is reviewed, debated, and summarised in this study, which begins with a brief overview of several graphene fabrication processes.

Table of Contents

1	Introduction	4
2	Aim and objective	4
3	Literature review.....	4
3.1	Preparation of graphene.....	5
3.1.1	Micromechanical exfoliation.....	5
3.1.2	Chemical vapour deposition	6
3.1.3	Liquid-phase exfoliation.....	6
3.1.4	Electrochemical synthesis	7
4	Microstructure of graphene.....	8
4.1	2D Graphene	8
4.1.1	Functionalized graphene sheet.....	9
4.1.2	Structure of catalyst loaded graphene	12
4.2	3D Graphene	13
4.2.1	Graphene aerogel	14
4.2.2	Graphene foam	16
5	Applications of the graphene.....	17
6	Conclusion.....	18
7	Reference	20

1 Introduction

Graphene is a material that has the potential to open up new industries and perhaps modify old techniques and equipment. Brahim Assa et al. highlighted the prospect of effectively integrating graphene directly into a device, allowing for a wide range of applications such as actuation, photovoltaics, thermoelectricity, shape memory, self-healing, electro-rheology, and space missions. Graphene's mass and low-cost manufacturing has become a critical step towards this real-world uses, through innovative academic and industrial research. [1]

Graphene is a carbon-based substance that is comparable to graphite but has properties that make it very light and strong. A sheet of graphene measuring one square metre weighs 0.78 milligrams. Its tensile strength is 195 times that of steel, and its density is comparable to carbon fibre. [2] All of this allows it to withstand large bending stress without cracking. It is one of the most electrically and thermally conductive materials, making it ideal for electronics and a variety of other sectors.

2 Aim and objective

Our aim is to understand the microstructure of graphene and study the areas of its future applications. We defined the following objectives to achieve our aim.

- Introduction to graphene and comparative analysis with recently developed smart materials.
- Characteristics of the microstructure of the graphene and characteristic of properties.
- Applications of the graphene

3 Literature review

Many graphene microstructure-engineering strategies have recently been developed, [3], such as 2D graphene sheets, 3D graphene interconnection networks, and their reconfigured derivatives, all of which play important and diverse roles in a variety of applications.

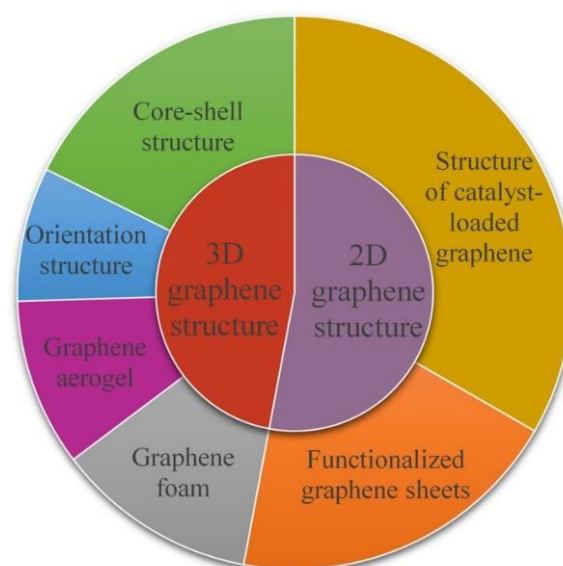


Figure 1 Different microstructures of graphene, Image credit: wileyonlinelibrary.com [4]

This paper focuses on graphene microstructure engineering's latest advancements. First, we will give a quick review of the various ways for obtaining graphene. Aside from that, these recent developments are thoroughly evaluated, debated, and summarised. Finally, we offer some perspectives on graphene microstructure engineering's future development plan and anticipated hurdles.

3.1 Preparation of graphene

In 2004, mechanical exfoliation on graphite was used for the first time to generate graphene. [5] The single or very few layer graphene with excellent structure may be obtained using this procedure. It is, however, both costly and inefficient. Numerous unique techniques to synthesize graphene have been proposed during the last 15 years, thanks to the efforts and discoveries of researchers.

3.1.1 Micromechanical exfoliation

Micromechanical exfoliation is a physical process for obtaining graphene from graphite by overcoming the weak connection between layers. For the first time, Geim et al. [6] used scotch tape bonding and separation to create monolayer graphene. (See Illustration 2) Dimiev et al. [7] improved their procedure after that. They used zinc film instead of scotch tape to create single-layer graphene using lithography and sputtering techniques. Micromechanical exfoliation has also been used to effectively manufacture single and few-layer graphene in other studies. [8-9]

Despite the fact that the micromechanical exfoliation approach can preserve graphene's exceptional characteristics, large-scale commercial manufacturing is problematic due to poor yield.

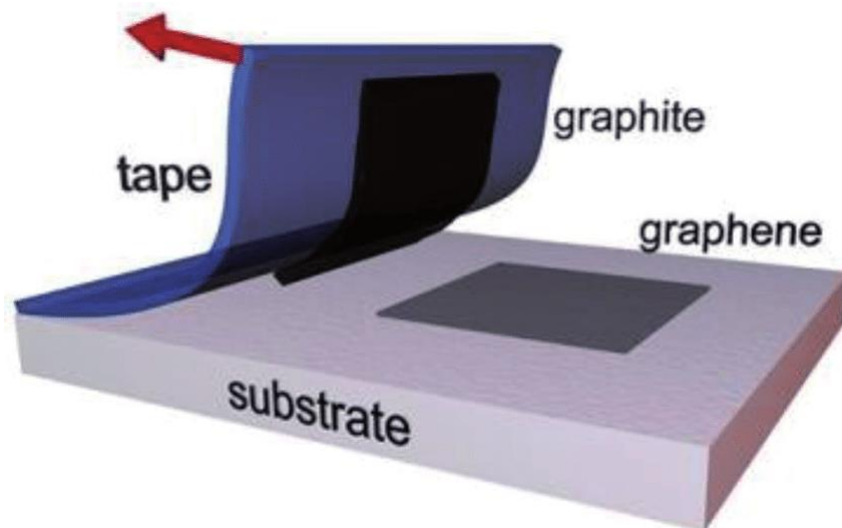


Figure 2 Micromechanical exfoliation process of Graphene, Image credit: Geim et al. [6]

3.1.2 Chemical vapour deposition

The "bottom-up" synthesis of large-area graphene using chemical vapour deposition (CVD) is a scalable and dependable approach. [10-12] Kim et al. [13] described the direct CVD creation of large-scale graphene sheets on thin nickel layers, as well as two techniques for patterning and transferring the films to other substrates (Figure 3). Consequently, the graphene developed had a high electron mobility and a low sheet resistance. Furthermore, comparable findings were obtained in other similar investigations. The high cost and strong process conditions limit the vast uses of CVD, much as a coin has two sides. [14-17]

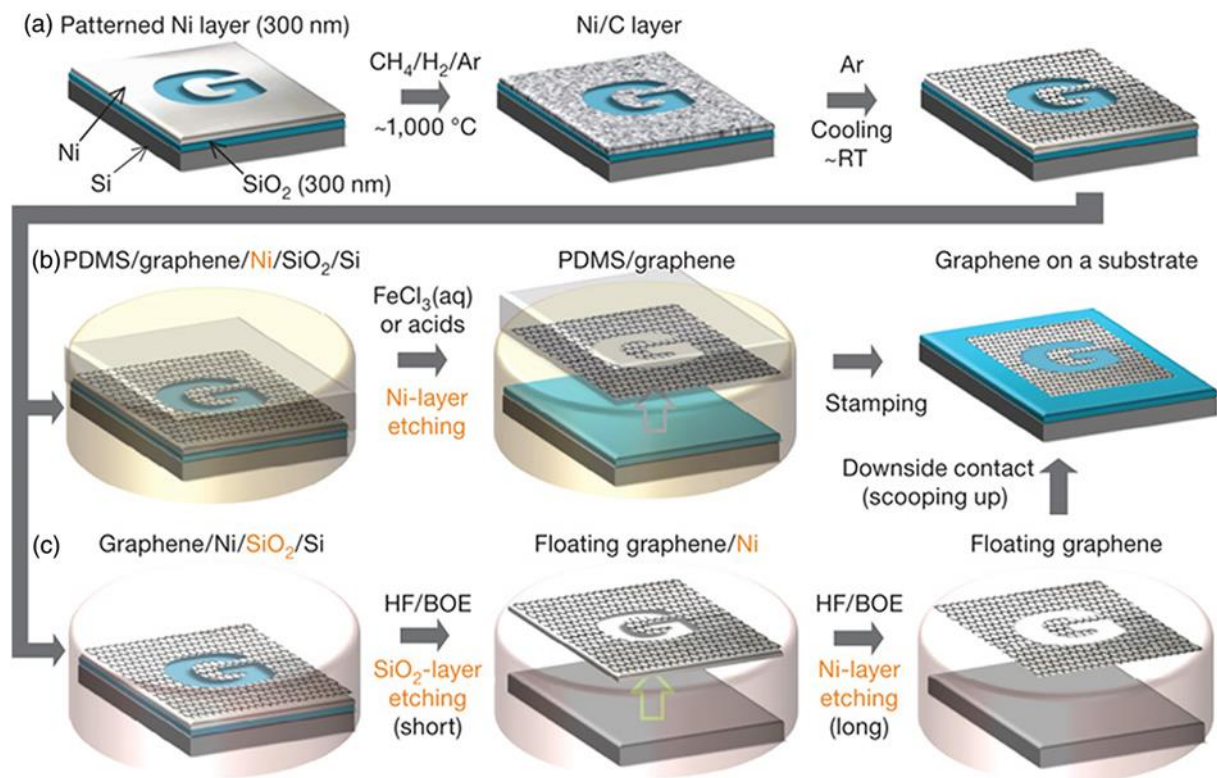


Figure 3 Chemical vapour deposition method for the preparation of Graphene, Image credit: Kim et al. [13]

3.1.3 Liquid-phase exfoliation

Solvents with surface energies that match those of graphene (such as DMF and NMP, for example) are employed to equalize the interaction between graphite layers in this approach. When their surfaces are aligned, the contact between them can balance a portion of the interlayer van der Waals force, allowing graphene dispersion to be produced following ultrasound, heating, or shear stress. [18] Hernandez et al. [19] used liquid-phase exfoliation to produce high-quality single layer graphene with a high yield.

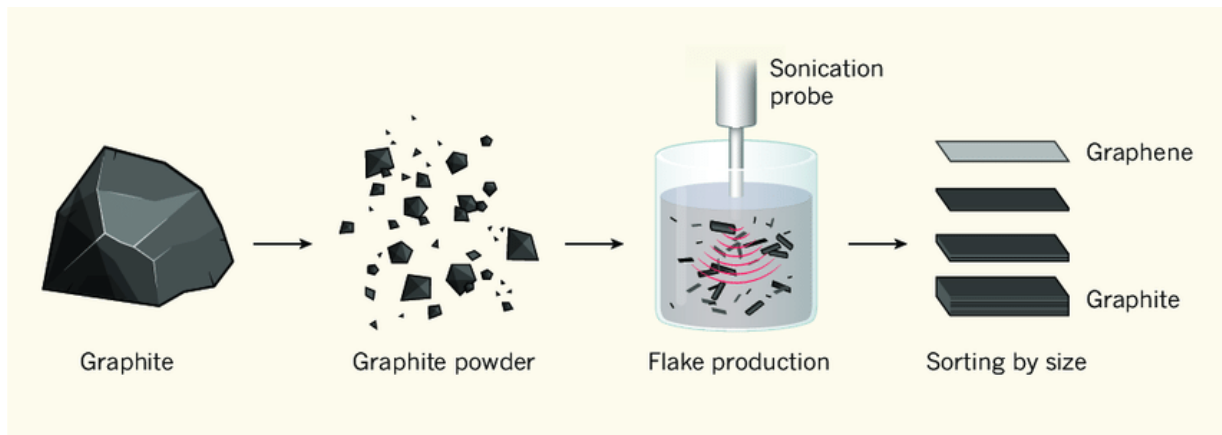


Figure 4 Liquid-phase exfoliation method for the preparation of Graphene, Image credit: Hernandez et al. [19]

Exfoliation in the liquid phase is mainly a physical process. Because there is no chemical reaction during the whole production process, graphene's exceptional characteristics and structure may be preserved. However, the hazardous organic solvents employed in the manufacturing process are rather difficult to extract, resulting in waste and contamination.

3.1.4 Electrochemical synthesis

In the realm of material preparation, electrochemical processes including metal deposition & metal oxide have been widely used. [20-21] and graphite is frequently utilised like an electrode material in the electrochemical area, setting the groundwork for graphene production via electrochemical means. Figure 5 depicts the electrochemical graphene peeling mechanism. [22-23] During the transesterification reaction, reduction happens at the cathode, which has a graphite electrode, and oxidation occurs at the anode, which is often gold or other metal elements.

The anions in the electrolyte are generally continually injected into the graphite layers, which reduces the van der Waals force between both the graphite sheets when the electric field is applied. Around the same time, the produced gases at the cathode has the potential to stimulate graphene exfoliation. Graphene would be separated from the electrode when the electrochemical process reaches a particular point, and graphene dispersion may then be obtained.

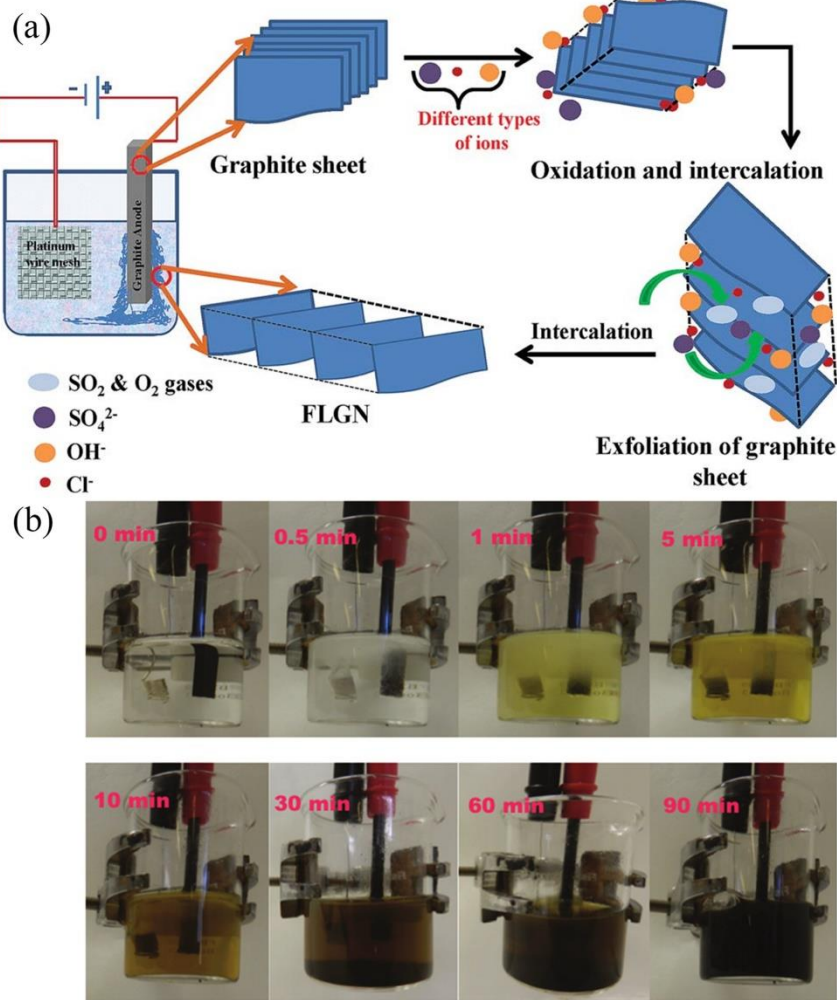


Figure 5 Electrochemical synthesis method for preparation of Graphene, Image credit: [22-23]

4 Microstructure of graphene

4.1 2D Graphene

Graphene represents a fundamentally new family of materials, known as two-dimensional (2D) materials, which are only one atom thick. They are named 2D because they only have two dimensions: length and breadth; because the substance is just single atom thick, the third dimension, elevation, is ignored. Furthermore, graphene's extraordinary capabilities are only visible at this single- or few-layer stage. Other graphitic materials, such as carbon nanotubes, are made up of graphene: Graphite is made up of carbon atoms bonded together; a carbon nanotube is made up of carbon atoms rolled together; and a fullerene is made up of carbon atoms sliced and folded into a spherical form. (Figure 6)

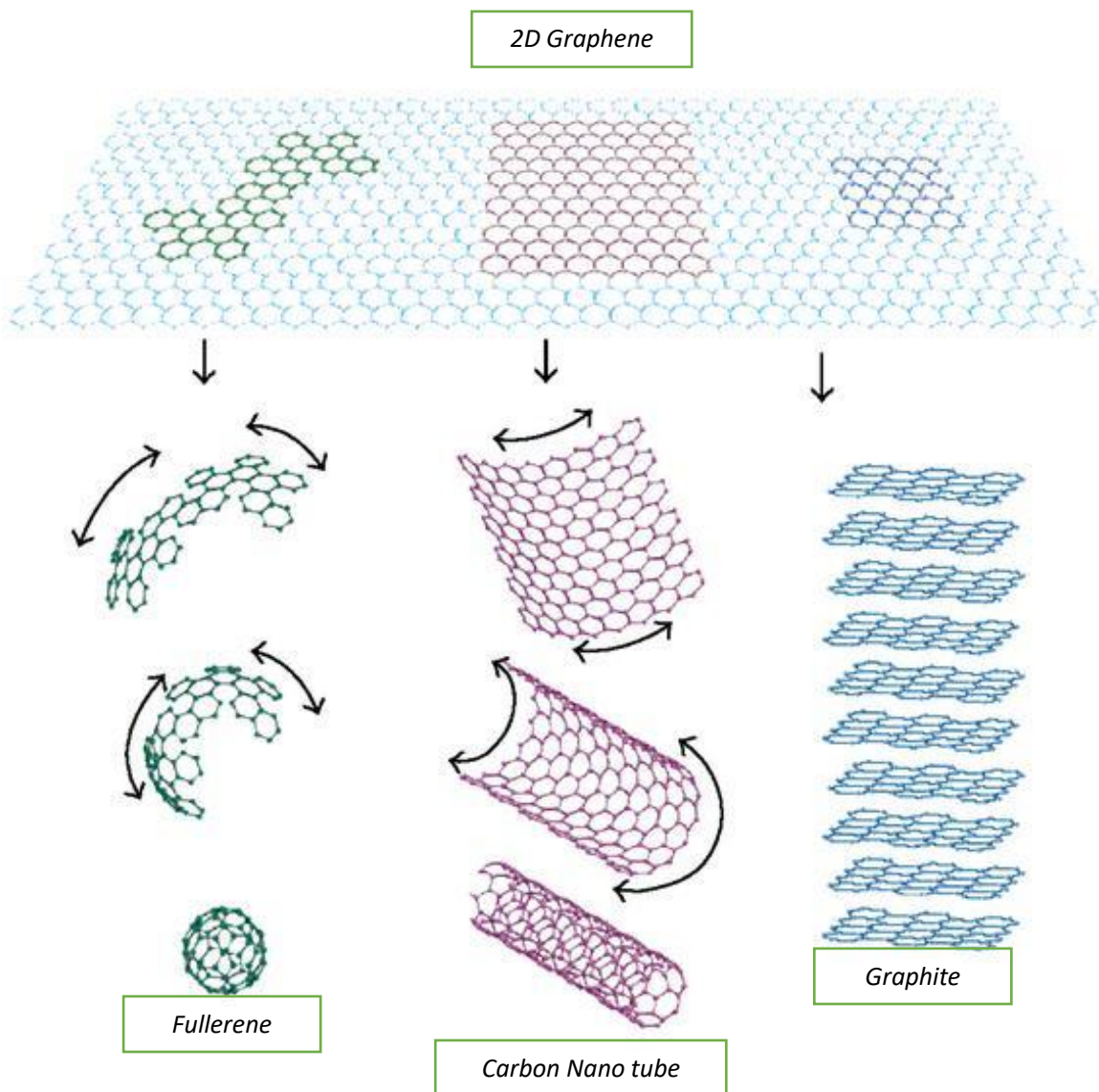


Figure 6 Graphene sheets as building blocks for other graphitic materials, Image credit: [Nat. Mater., 6:183–191, 2007]

Graphene has sparked interest because of its remarkable two-dimensional structure and application possibilities. Graphene is a best choice to be used in nanoelectromechanical systems due to its remarkable strength and low mass density, that are of considerable interest for both fundamental research of dynamics at the nanometre scale and for a variety of applications such as coercion, position, mass, and gas sensing [24].

4.1.1 Functionalized graphene sheet

Figure 6 depicts a graphene sheet with an interconnected and persistent six-membered ring structure. Strong intermolecular forces (such as π - π interactions) between graphene sheets are aided by the interconnected and persistent six-membered ring structure. As a result, graphene sheets tend to stack together. The chemically inert exterior and insolubility in

solvent of agglomerated graphene sheets make creating inorganic composite materials extremely challenging.

Thus, well-dispersed graphene sheets are required to provide outstanding composite performance. As a result, it is critical to increase the compatibility of graphene sheets in varied settings. Many related studies and investigations have been conducted to now.

One of the most common approaches for modifying graphene sheets is functionalization [25]. There are two types of functionalization methods: covalent and non-covalent. Covalent functionalization methods create covalent links between graphene sheets, and non-covalent methods add non-covalent bonds (such as ionic bonds) between graphene sheets.

Covalent bonds are easily formed at graphene borders and/or surface imperfections with high activity [26]. Through acidizing treatment, the methods create hydrophilic oxygen-containing groups, and graphene sheets might be further functionalized by adding new groups or diverse molecule chains through reactivity with oxygen-containing groups.

Yang et al. [27], for example, covalently functionalized using amine-terminated ionic liquid without the use of polymeric or surfactant stabilisers. (See Figure 7) The resultant functionalized graphene sheets diffused efficiently in water, N,N-dimethyl-formamide, and dimethyl sulfoxide, in that order.

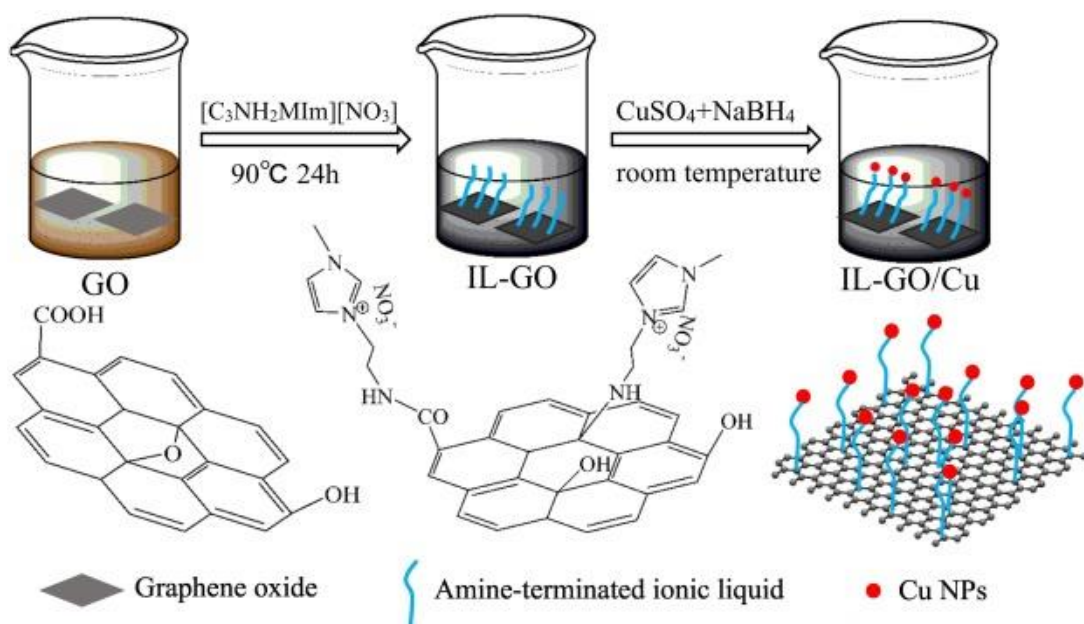


Figure 7 Covalent functionalization with amine-terminated ionic liquid, Image credit: Yang et al. [27]

Wang et al. [28] created aqueous graphene-poly sodium 4-styrene sulfonate by covalently functionalizing graphene oxide with poly sodium 4-styrene sulfonate and reducing it with hydrazine hydrate under hydrothermal conditions. (See Figure 8) They also created organophilic graphene octadecylamine by reacting it with octadecylamine and then reducing it with hydroquinone in a reflux procedure.

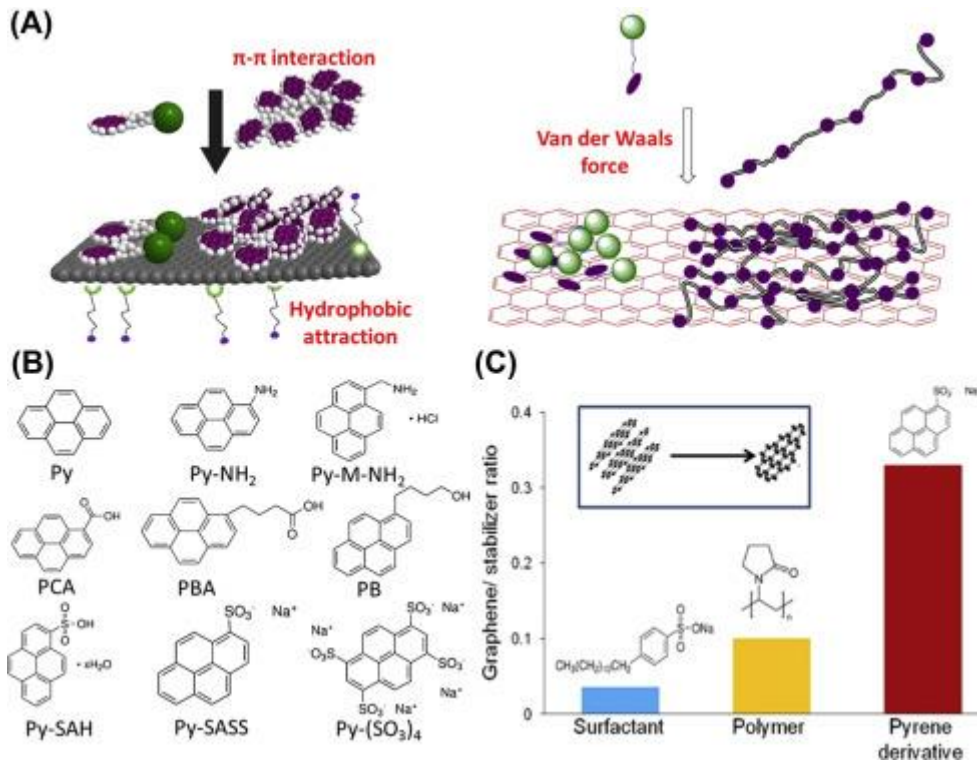


Figure 8 Covalent functionalization: graphene oxide with poly sodium 4-styrene sulfonate, Image credit: Wang et al. [28]

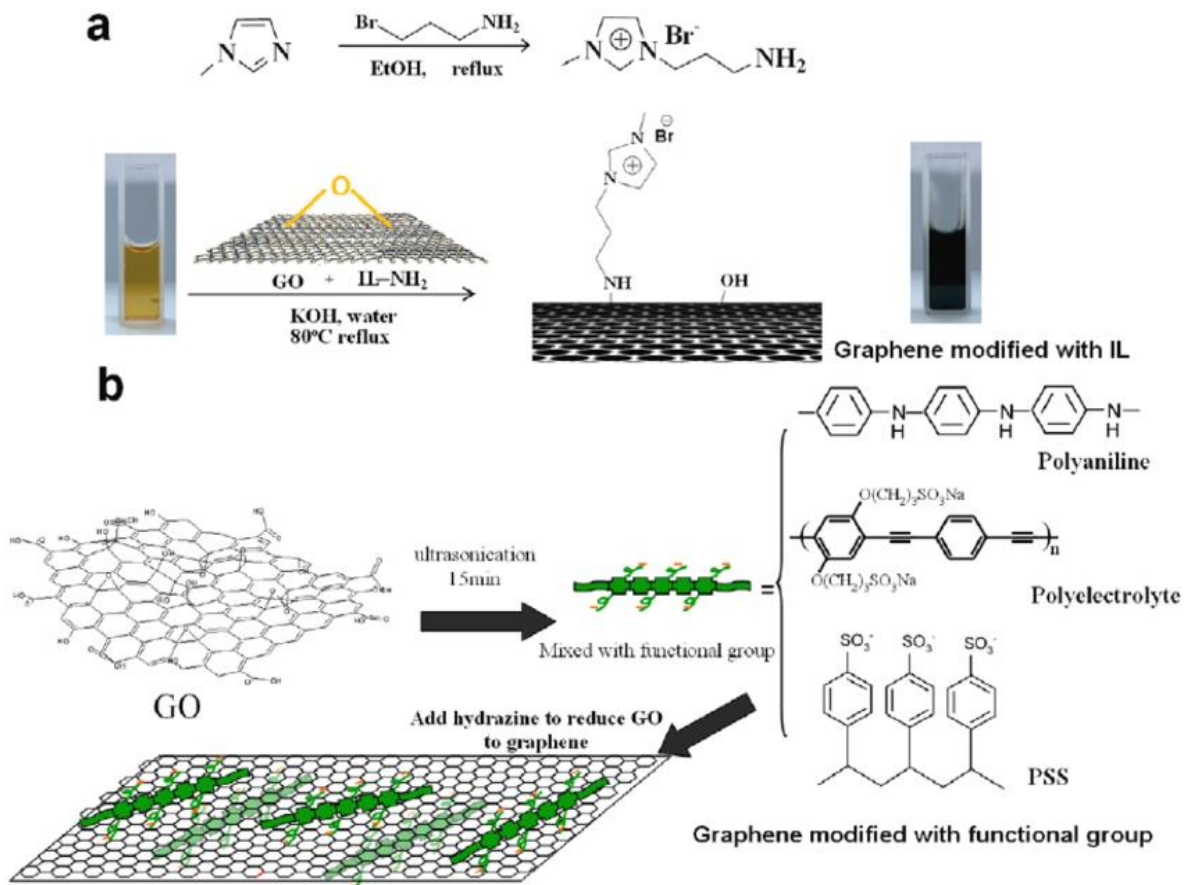


Figure 9 Non-covalent functionalization, Image credit: Park et al. [29]

Non-covalent functionalization, on the other hand, is mostly dependent on molecule interaction, and the alteration of molecule and ion allows graphene to be distributed in solvents stably. For example, Park et al. [29] employed the strong base KOH to impart a significant negative charge on graphene oxide via interactions with strong carboxylic acid and epoxy groups, leading to reduced graphene oxide that can be stably distributed in water for a period 3 months. (See Figure 9)

Covalent functionalization cannot avoid graphene structural degradation, and there are often some remaining groups on the surface, which affects graphene performance even when treated with heat reduction. Because of graphene's integrated structure and superior performance, non-covalent method rely on inter molecular are preferable to covalent ones.

According to Xu et al. [30], a stable aqueous dispersion of graphene sheets generated by graphene oxide reduction may be non-covalently functionalized with such a water-soluble pyrene derivative by π -stacking. The functionalized graphene sheet film had a high conductivity based on this dispersion, which was seven orders of magnitude more than that of a graphene oxide film.

4.1.2 Structure of catalyst loaded graphene

Graphene-based single-atomic catalysts are becoming increasingly important in a variety of uses, particularly renewable energy transformation and environmental systems. This synergistic structure, which typically consists of atomic metal active sites stabilised on a strong graphene support, is having broken through the short comings of important and valuable catalysts and expanding the range of applications while achieving high atom utilisation, phenomenal innovative catalytic activity, as well as conformational flexibility. (31–33)

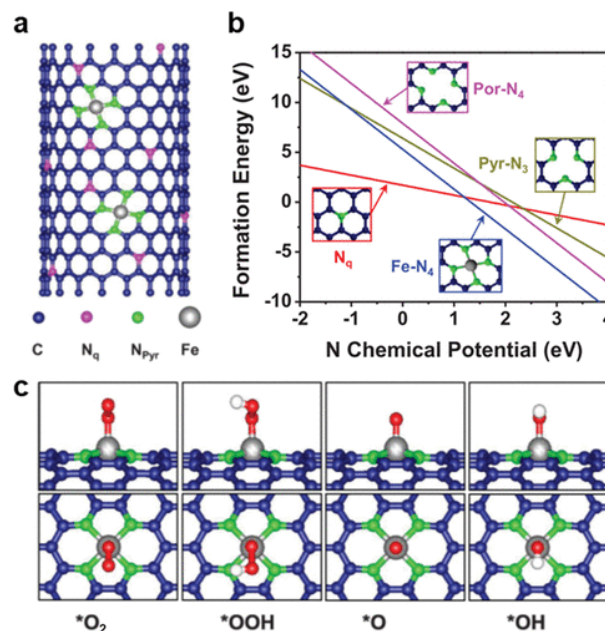


Figure 10 Formation energies as a function of the N chemical potential, Image credit: [35]

Selection of support material is also necessary to properly leverage the combined advantages of optimum catalyst. The arrangement of chemical coordination in the support material has a significant impact on the electronic structure of single-atomic catalyst sites. As a result, the catalytic activity of single-atomic catalysts is heavily reliant on the nature of the bonding between the catalytic single atoms and the support materials. (34)

Because of its unique features, such as high electrical conductivity and huge surface area, graphene has been identified as an appropriate support material for electrochemical catalysts. (35–37) Unfortunately, keeping the benefits of the many anchoring sites while maintaining the real chemically inert surface of pure graphene necessitates sophisticated chemical modification. (Figure 10) In the early stages of carbon-based catalyst research, the transition metal–nitrogen (N) moiety was postulated for catalytic activity activation without a systematic knowledge of the role of dopants. (38–40)

4.2 3D Graphene

Graphene has long piqued the interest of metallurgists, but in its most basic form, it is made up of 2D flat sheets that are only an atom thick yet may theoretically expand in width and length forever. Graphene must be coaxed into a three-dimensional shape before it can be used as an engineering material, but thus far, attempts have been many magnitudes weaker than projected.

Graphene's two-dimensional nature makes it similar to the laser in the 1960s in terms of being an answer in search of a problem. A team of MIT scientists, led by David L. Chandler et al. [41], has invented a sponge-like 3D form of graphene that has just 5% of the density of steel yet is 10 times as strong.

The researchers discovered that by compressing microscopic layers of graphene under pressure and temperature, they could produce robust, stable porous structures with a large high surface to volume ratio, akin to coral. These configurations, according to the researchers, allow two-dimensional graphene to create robust structure in the same manner that sheets of paper could be folded and rolled into considerably stronger forms, such as cylinders and corrugations, that can withstand significant weights.

Using this as a preliminary step, high-resolution 3D-printed models of various shapes were created out of plastic – akin to the "nerf-like" porous materials called gyroids that graphene naturally forms, but thousands of times larger. These forms are so complicated, according to David L. Chandler et al., that printing is the only realistic technique to construct them. The tensile strength and compression resistance of these geometries were then tested, and the results were compared to the computer calculations. (See Figure 11).

The experiments revealed that graphene in a three-dimensional form had a density of 5% that of steel, but 10 times the strength. The researchers discovered that much of this was due to

the geometrical arrangement rather than the substance. Similar improvements in strength were observed when graphene was replaced with polymers or metals.

According to David L. Chandler et al., the approach might be used to create 3D graphene structures in a variety of materials varying from polymer to structural concrete, resulting in constructions that are not only stronger and lighter, but also have greater insulating qualities. Furthermore, the porous materials might be employed in water or chemical plant filtration systems.

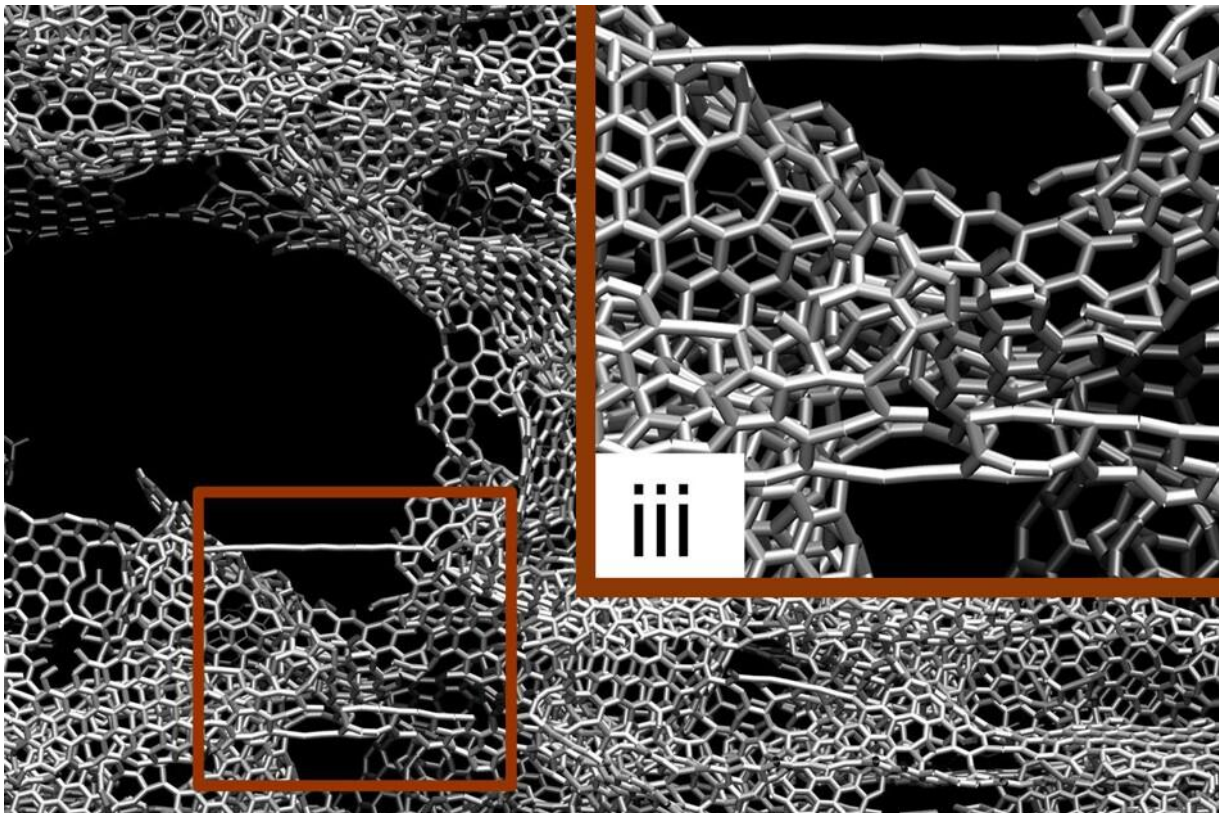


Figure 11 Simulation results of tensile and compression tests on 3-D graphene, Image credit: David L. Chandler et al. [41]

4.2.1 Graphene aerogel

The way graphene aerogels are constructed has a big impact on their physicochemical characteristics. Regardless as to whether the bridge between both the graphene layers are physical or chemical, mechanical characteristics can vary dramatically. Low - impedance connections among graphene sheets are required for electrical characteristics. Electrochemically active or catalytic compounds can be used to increase functionality. The next part will look at how different components can affect the structure and functioning of graphene aerogel by introducing them into the carbon matrix.

The resorcinol-formaldehyde (RF) technique for manufacturing carbon aerogels was published in 1989 as one of the most essential, affordable, and practical synthetic ways for obtaining graphene aerogel. [42] Worsley et al. created 3D graphene assemblies in 2011 by varying the quantities of RF reactants in an 11 mg mL⁻¹ graphene oxide solution. [43] Lim et al. used hydrochloric acid (HCl) as a catalytic and acetonitrile as the solvent to crosslink RF and graphene oxide, reducing the gelation period of graphene from several days to 1–2 hours. (See Figure 12). [44]

Scaffaro et al. recently developed a lightweight graphene-based aerogel by combining graphene oxide with an amino terminal polyethylene glycol (PEGNH₂) in an aqueous environment using carbodiimide (EDC), then freeze-drying it (Figure 13). [45]

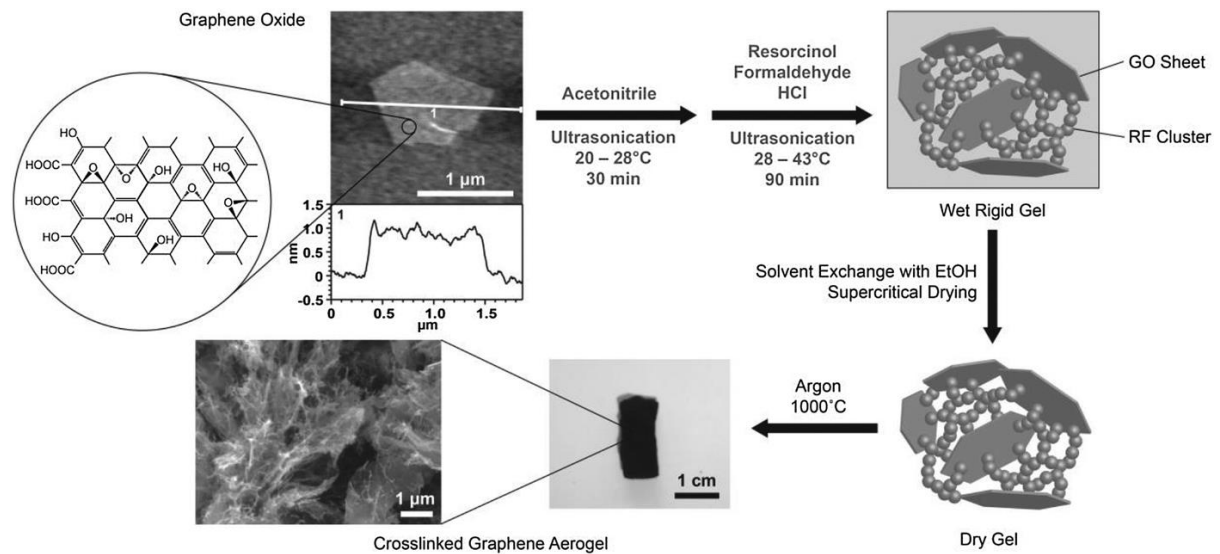


Figure 12 Diagram of the Graphene oxide- Resorcinol Formaldehyde aerogel preparation process, Image credit: Lim et al. [44]

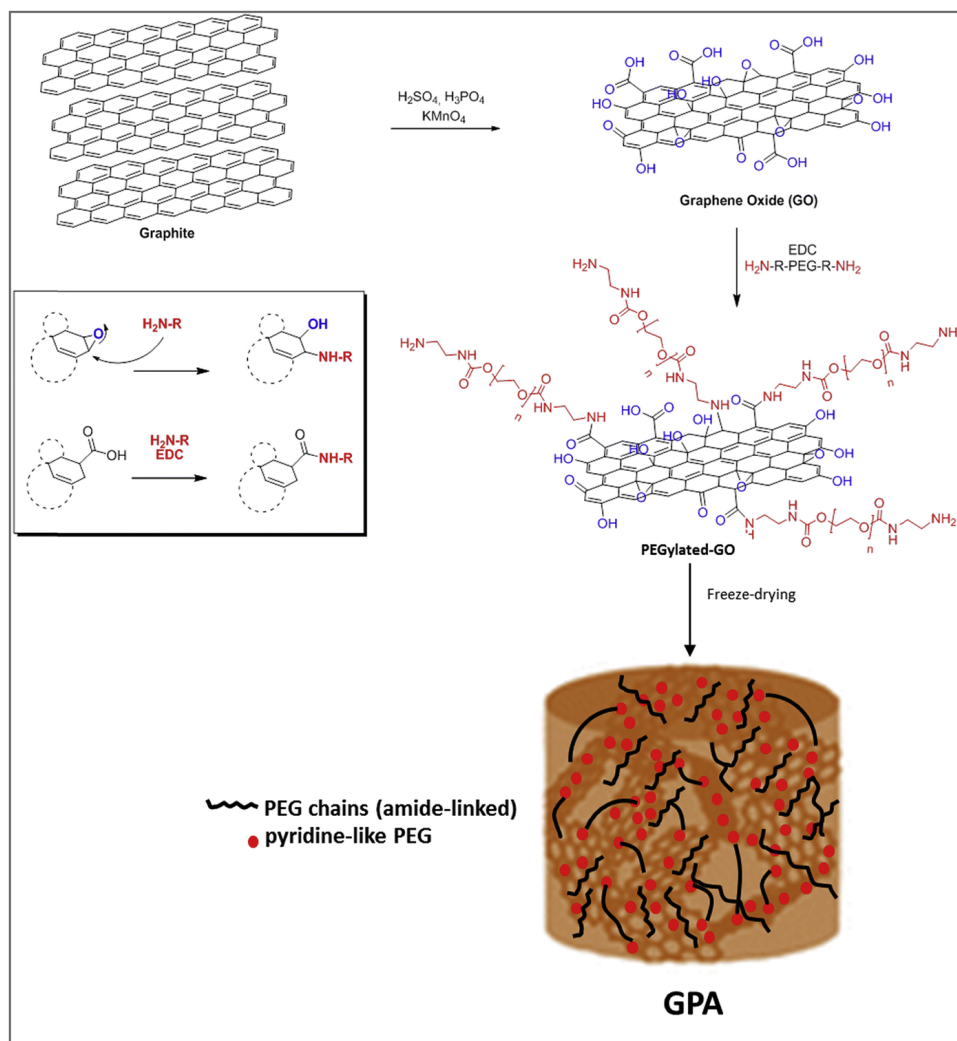


Figure 13 Schematics of the pathway followed to synthesize GO-PEG aerogel (GPA), Image credit: Scaffaro et al. [45]

From the standpoint of sustainable development, the development of sophisticated multifunctional materials for economic and carbon concerns is becoming increasingly significant. Graphene aerogels exhibit exceptional nanoscale effects because of their structural stability and interconnected porosity, resulting in new composite materials with superior characteristics and innovative functions. The logical adjustment of the materials characteristics of such graphene-based 3D macrostructures is the key to improving their energy performance, thanks to advancements in processing techniques. The exceptional capabilities of these novel materials will spur the development of new technologies in areas such as energy storing, filtration and separation, catalysis, and sensors, among others.

4.2.2 Graphene foam

Because of their unique features, 3D graphene foams are a good choice for providing circumstances that resemble *in vivo* settings, allowing for successful cell adhesion, proliferation, and differentiation. The highest biocompatibility among nanostructures, a high surface-to-volume ratio, a 3D porous structure (to allow for homogeneous/isotropic tissue

growth), highly favourable mechanical properties, and rapid mass and electron transport kinetics (that are all considered necessary for chemical/physical activation of differentiated cells) are among them.

Chen et al. used a template approach (this method uses supramolecular assemblages such as microspheres and emulsification as a template, [46]) to deposit 3D graphene sheets on an interlinked 3D macroporous architecture of Ni foam by pyrolyzing CH₄ at 1000 °C as the first attempt. [46] They placed a coating of poly (methyl methacrylate) on the surface of a Ni template to prevent the graphene network from collapsing during the etching procedure in HCl solution. Finally, they utilised hot acetone to remove the poly (methyl methacrylate) layer after etching the Ni template. (Figure 14) [47].

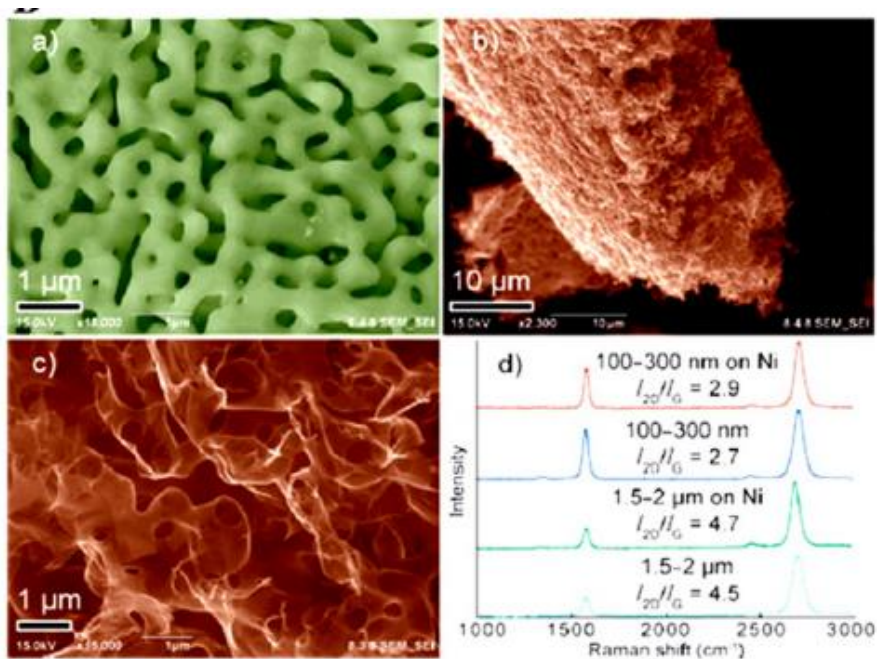


Figure 14 SEM images of Graphene foam, 3D graphene on nonporous Ni template, Image credit: Chen et al. [47]

5 Applications of the graphene

The efficiency of batteries and super capacitors, according to Fen & Jiang et al., is dependent on the concentration at that they can store electricity and the frequency at which they'll be charged and discharged. The nanostructured electrodes utilised in these energy systems are important to these functionalities. Carbon nanomaterials with pores are commonly used as electrodes in super capacitors and commercialized lithium ion batteries. Porous carbon, such as activated carbon, microporous carbon, and highly porous carbon, has a large surface area and a tuneable porous structure, yet it has a low electrical conductivity. Graphene, on the other hand, has an excellent electrical conductivity but a small surface area. It is feasible to construct conductivity scaffold to host sulphur for lithium ion storage batteries by building a 3D graphene framework. [48]

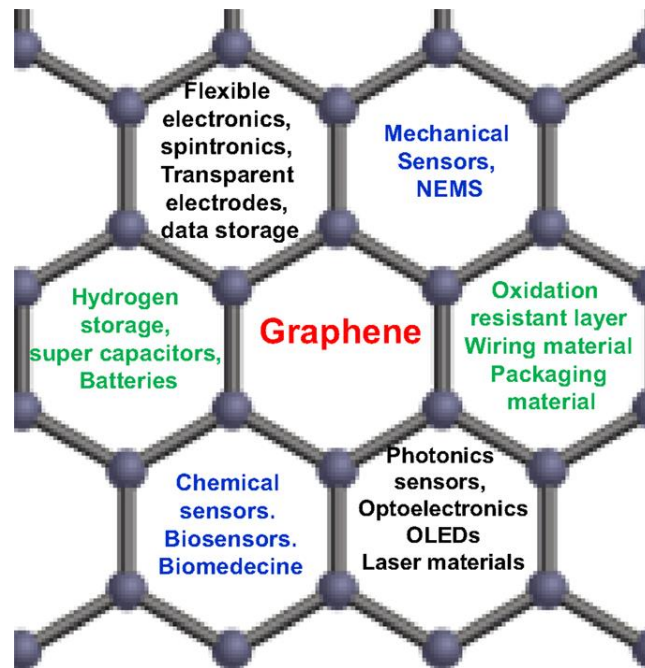


Figure 15 Potential applications of graphene, Image credit: Fen & Jiang et al. [48]

Xiangru Song et al. (2018) discovered better power generation and compelling water filtration in microbial energy components using graphene-based film air-cathodes, and said that graphene-based cathodes may generate electricity and had reduced cathode bio fouling. To improve the presentation of MFC and provide a sifting profluent, they used a stage reversal technique to produce graphene-containing air-cathode film. [49]

Houk Jang et al. (2016) looked at recent research on graphene-based stretchy and adaptive devices. Contact circuits, Nano producers, controllers, biomaterials, and solar cell were among the applications that shown great pertinence. The final results proved graphene's enormous potential as a component in flexible and adaptive devices. [50]

Kena Chen et al. (2017) looked at the challenges and potential of graphene-based materials for flexible energy storage devices. Adaptable graphene-based two-dimensional sheet and one-dimensional fibres super capacitors, as well as lithium-ion, lithium–sulphur, and other batteries, were on display. [51]

Hong et al. (2018) investigated and concluded that adding graphene to a geopolymers improves its characteristics. The results revealed that with the inclusion of graphene, the cementitious products were denser and the inner porosity was decreased. [52]

6 Conclusion

Graphene is the first two-dimensional atomic crystal created in a laboratory. Graphene is described as an alternative to many traditional materials in various applications, with excellent physical and chemical properties such as mechanical stiffness, strength, and elasticity, as well as remarkably high electrical and thermal conductivity. A sheet of graphene measuring one square meter weighs 0.78 milligrams. Its tensile strength is 195 times that of steel, and its density is comparable to carbon fiber. In this paper, we gave a quick review of the various ways for obtaining graphene. Aside from that, these recent developments are

thoroughly evaluated and summarized. We discussed the 2D and 3D graphene microstructure, methods to prepare 2D and 3D graphene and properties of the same graphene material. In 2D graphene, we discussed functionalized graphene sheet and structure of catalyst-loaded graphene. In 3D graphene, we discussed the graphene aerogel and graphene foam.

The microstructure of graphene differs significantly depending on the method of 3D graphene synthesis. The template restricts the porosity and specific surface area of graphene foam. It has a greater cost and a more complicated preparation procedure than graphene aerogel. The orientation arrangement may fully use graphene's in-plane thermal conductivity while also considerably improving its mechanical characteristics.

7 Reference

1. Aïssa, B., Memon, N.K., Ali, A. and Khraisheh, M.K. (2015). Recent Progress in the Growth and Applications of Graphene as a Smart Material: A Review. *Frontiers in Materials*, 2.
2. Nanografi (2019). 60 Uses of Graphene – The Ultimate Guide to Graphene’s (Potential) Applications in 2019. [online] Nanografi Nano Technology. Available at: <https://nanografi.com/blog/60-uses-of-graphene/>.
3. Hossain, S. A., Hossen, M., & Anower, S. (2018). ESTIMATION OF DAMSELFISH BIOMASS USING AN ACOUSTIC SIGNAL PROCESSING TECHNIQUE. *Journal of Ocean Technology*, 13(2).’
4. Wiley.com. (2019). Wiley Online Library | Scientific research articles, journals, books, and reference works. [online] Available at: <https://onlinelibrary.wiley.com/>.
5. Liu, Y., Zheng, J., Zhang, X., Li, K., Du, Y., Yu, G., Jia, Y. and Zhang, Y. (2021). Recent advances on graphene microstructure engineering for propellant-related applications. *Journal of Applied Polymer Science*, 138(21), p.50474.
6. Novoselov, K. S., Geim, A. K., Morozov, S. V., Jiang, D., Zhang, Y., Dubonos, S. V., Grigorieva, I. V., & Firsov, A. A. (2004). Electric field effect in atomically thin carbon films. *Science (New York, N.Y.)*, 306(5696), 666–669. <https://doi.org/10.1126/science.1102896>.
7. Gunlycke, D., & Sheehan, P. E. (2011). Local Peeling of Graphene. *Science*, 331(6021), 1146–1147. <https://doi.org/10.1126/science.120077>.
8. Zhao, W., Fang, M., Wu, F., Wu, H., Wang, L. and Chen, G. (2010). Preparation of graphene by exfoliation of graphite using wet ball milling. *Journal of Materials Chemistry*, 20(28), p.5817..
9. Y. Fan, L. Wang, J. Li, J. Li, S. Sun, F. Chen, L. Chen, W. Jiang, *Carbon* 2010, 48, 1743.
10. Muñoz, R. and Gómez-Aleixandre, C. (2013). Review of CVD Synthesis of Graphene. *Chemical Vapor Deposition*, 19(10-11-12), pp.297–322.
11. Scirp.org. (2016). Somani, P.R., Somani, S.P. and Umeno, M. (2006) Planer Nano-Graphenes from Camphor by CVD. *Chemical Physics Letters*, 430, 56-59. - References - Scientific Research Publishing. [online] [Accessed 20 Apr. 2022].
12. Z. Chen, W. Ren, B. Liu, L. Gao, S. Pei, Z.-S. Wu, J. Zhao, H.-M. Cheng, *Carbon* 2010, 48, 3543.
13. Kim, K. S., Zhao, Y., Jang, H., Lee, S. Y., Kim, J. M., Kim, K. S., Ahn, J.-H., Kim, P., Choi, J.-Y., & Hong, B. H. (2009). Large-scale pattern growth of graphene films for stretchable transparent electrodes. *Nature*, 457(7230), 706–710. <https://doi.org/10.1038/nature07719>.
14. Sun, Z., Yan, Z., Yao, J., Beitler, E., Zhu, Y. and Tour, J.M. (2011). Erratum: Growth of graphene from solid carbon sources. *Nature*, 471(7336), pp.124–124.
15. Z. Yan, Z. Peng, Z. Sun, J. Yao, Y. Zhu, Z. Liu, P. M. Ajayan, J. M. Tour, *ACS Nano* 2011, 5, 8187.
16. Y. Zhang, T. Gao, S. Xie, B. Dai, L. Fu, Y. Gao, Y. Chen, M. Liu, Z. Liu, *Nano Res.* 2012, 5, 402.
17. P. Xu, J. Kang, J.-B. Choi, J. Suhr, J. Yu, F. Li, J.-H. Byun, B.-S. Kim, T.-W. Chou, *ACS Nano* 2014, 8, 9437.

18. W. S. Hummers, R. E. Offeman, *J. Am. Chem. Soc.* 1958, 80, 1334.
19. Y. Hernandez, V. Nicolosi, M. Lotya, F. M. Blighe, Z. Sun, S. De, I. T. McGovern, B. Holland, M. Byrne, Y. K. Gun'ko, J. J. Boland, P. Niraj, G. Duesberg, S. Krishnamurthy, R. Goodhue, J. Hutchison, V. Scardaci, A. C. Ferrari, J. N. Coleman, *Nat. Nanotechnol.* 2008, 3, 563.
20. M. Z. Hu, C. E. Easterly, *Mater. Sci. Eng., C* 2009, 29, 726.
21. J. García, M. Méndez, S. González, V. Vega, R. Caballero, V. M. Prida, in *Magnetic Nano- and Microwires*, 2nd ed. (Ed: 20 of 22 LIU ET AL. M. Vázquez), Woodhead Publishing, Swaston, Cambridge 2020.
22. N. Parveen, M. O. Ansari, M. H. Cho, *RSC Adv.* 2015, 5, 44920.
23. A. M. Abdelkader, I. A. Kinloch, R. A. W. Dryfe, *Chem. Commun.* 2014, 50, 8402.
24. B. Sun and X. M. Henry Huang, "Mechanical nano-resonators at ultra-high frequency and their potential applications," *South Africa Journal of Science*, vol. 104, pp. 169–171, 2008.
25. Park S, Ruoff R S. Chemical methods for the production of graphenes. *Nat Nanotech*, 2009, 4: 217–224
26. Boukhvalov D W, Katsnelson M I. Chemical functionalization of graphene with defects. *Nano Lett*, 2008, 8: 4373–4379
27. Yang H, Shan C, Li F, et al. Covalent functionalization of polydisperse chemically-converted graphene sheets with amine-terminated ionic liquid. *Chem Commun*, 2009, 26: 3880–3882
28. Wang G, Shen X, Wang B, et al. Synthesis and characterisation of hydrophilic and organophilic graphene nanosheets. *Carbon*, 2009, 47: 1359–1364
29. Park S G, An J, Piner R D, et al. Aqueous suspension and characterization of chemically modified graphene sheets. *Chem Mater*, 2008, 20: 6592–6594
30. Xu Y, Bai H, Lu G, et al. Flexible graphene films via the filtration of water-soluble noncovalent functionalized graphene sheets. *J Am Chem Soc*, 2008, 130: 5856–5857
31. Yang, X.-F.; Wang, A.; Qiao, B.; Li, J.; Liu, J.; Zhang, T. Single-atom catalysts: a new frontier in heterogeneous catalysis. *Acc. Chem. Res.* 2013, 46 (8), 1740– 1748.
32. Liu, J. Catalysis by supported single metal atoms. *ACS Catal.* 2017, 7 (1), 34– 59.
33. Wang, A.; Li, J.; Zhang, T. Heterogeneous single-atom catalysis. *Nat. Rev. Chem.* 2018, 2 (6), 65– 81.
34. Fei, H.; Dong, J.; Chen, D.; Hu, T.; Duan, X.; Shakir, I.; Huang, Y.; Duan, X. Single atom electrocatalysts supported on graphene or graphene-like carbons. *Chem. Soc. Rev.* 2019, 48 (20), 5207– 5241.
35. Wang, H.; Casalongue, H. S.; Liang, Y.; Dai, H. Ni(OH)₂ nanoplates grown on graphene as advanced electrochemical pseudocapacitor materials. *J. Am. Chem. Soc.* 2010, 132 (21), 7472– 7477.
36. Gawande, M. B.; Fornasiero, P.; Zbořil, R. Carbon-Based Single-Atom Catalysts for Advanced Applications. *ACS Catal.* 2020, 10 (3), 2231– 2259.

37. Liang, Y.; Li, Y.; Wang, H.; Zhou, J.; Wang, J.; Regier, T.; Dai, H. Co₃O₄ nanocrystals on graphene as a synergistic catalyst for oxygen reduction reaction. *Nat. Mater.* 2011, 10 (10), 780–786.
38. Lefèvre, M.; Proietti, E.; Jaouen, F.; Dodelet, J.-P. Iron-based catalysts with improved oxygen reduction activity in polymer electrolyte fuel cells. *Science* 2009, 324 (5923), 71–74.
39. Bashyam, R.; Zelenay, P. A class of non-precious metal composite catalysts for fuel cells. *Materials for sustainable energy: A collection of peer-reviewed research and review articles from Nature Publishing Group; World Scientific*, 2011; pp 247–250
40. Gong, K.; Du, F.; Xia, Z.; Durstock, M.; Dai, L. Nitrogen-doped carbon nanotube arrays with high electrocatalytic activity for oxygen reduction. *Science* 2009, 323 (5915), 760–764.
41. Chandler, D.L. (2017). Researchers design one of the strongest, lightest materials known. [online] MIT News. Available at: <https://news.mit.edu/2017/3-d-graphene-strongest-lightest-materials-0106>.
42. “Reversible Polymeric Gels and Related Systems”, edited by P. S. Russo, ACS Symposium Series 350 (American Chemical Society, Washington, DC. 1987).
43. Chandrasekaran, S., Campbell, P.G., Baumann, T.F. et al. Carbon aerogel evolution: Allotrope, graphene-inspired, and 3D-printed aerogels. *Journal of Materials Research* 32, 4166–4185 (2017). <https://doi.org/10.1557/jmr.2017.411>
44. Matthew B. Lim, Matthew Hu, Sandeep Manandhar, Avery Sakshaug, Adam Strong, Leah Riley, Peter J. Pauzauskie, Ultrafast sol–gel synthesis of graphene aerogel materials, *Carbon*, Volume 95, 2015, Pages 616-624, ISSN 0008-6223, <https://doi.org/10.1016/j.carbon.2015.08.037>.
45. Roberto Scaffaro, Andrea Maio, Francesco Lopresti, Daniele Giallombardo, Luigi Botta, Maria Luisa Bondi, Simonpietro Agnello, Synthesis and self-assembly of a PEGylated-graphene aerogel, *Composites Science and Technology*, Volume 128, 2016, Pages 193-200, ISSN 0266-3538, <https://doi.org/10.1016/j.compscitech.2016.03.030>.
46. Huang, X.; Sun, B.; Su, D.; Zhao, D.; Wang, G. Soft-template synthesis of 3D porous graphene foams with tunable architectures for lithium–O₂ batteries and oil adsorption applications. *J. Mater. Chem. A* 2014, 2 (21), 7973–7979.
47. Chen, Z.; Ren, W.; Gao, L.; Liu, B.; Pei, S.; Cheng, H.-M. Threedimensional flexible and conductive interconnected graphene networks grown by chemical vapour deposition. *Nat. Mater.* 2011, 10 (6), 424.
48. Li, Fen & Jiang, Xue & Zhao, Jijun & Zhang, Shengbai. (2015). Graphene oxide: A promising Nanomaterial for energy and environmental applications. *Nano Energy*. 16. [10.1016/j.nanoen.2015.07.014](https://doi.org/10.1016/j.nanoen.2015.07.014).
49. Xiangru Song, Jia Liu, Qing Jiang, Youpeng Qu, Weihua He, Bruce E. Logan, Yujie Feng, Enhanced electricity generation and effective water filtration using graphene-based membrane air-cathodes in microbial fuel cells. *Journal of Power Sources*, Volume 395, 15 August 2018, Pages 221-227, DOI: [10.1016/j.jpowsour.2018.05.043](https://doi.org/10.1016/j.jpowsour.2018.05.043).
50. Houk Jang, Yong Ju Park, Xiang Chen, Tanmoy Das, Min-Seok Kim and Jong-Hyun Ahn. Graphene-Based Flexible and Stretchable Electronics. *Adv. Mater.* 2016, 28, 4184–4202, DOI: [10.1002/adma.201504245](https://doi.org/10.1002/adma.201504245).

51. Kena Chen, Qingrong Wang, Zhiqiang Niu, Jun Chen. Graphene-based materials for flexible energy storage devices. *Journal of Energy Chemistry*, Volume 27, Issue 1, 1 January 2018, Pages 12-24.
52. Hong, X., Lee, J.C. and Qian, B. (2022). Mechanical Properties and Microstructure of High-Strength Lightweight Concrete Incorporating Graphene Oxide. *Nanomaterials*, 12(5), p.833.