

The graphene and graphite landscape: Indications of unexplored territory

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Abstract

Graphene can be made by chemical vapour deposition (CVD) methods. CVD technology has been developing rapidly, and graphene can now be made in large sheets at scales of metres and speeds of a metre per minute. More than ten layers of graphene is considered to be graphite and, by implication, of less scientific and commercial interest. In contrast, a homogeneous structure of single crystal graphene stacks has not been studied. It will soon be possible to manufacture such Van der Waals (VdW) homostructures of multi-layer single crystal large-area sheet graphene at industrial scale.

This paper aims to raise awareness of the properties of the new multi-layered material because there are indications of impressive physical capabilities and commercial potential. Technically this is graphite, but in 'perfect' form. It may need a new name to distinguish it as a new frontier material.

Keywords: Frontier material, Graphene, Graphene homostructure, Graphite, Multi-layer single crystal graphene, Van der Waals homostructure.

1. Introduction

As Dreyer, Ruoff and Bielawski, (2010) noted, graphene chemistry has a rich history that can be traced as far back as 1840 [1]. Today, graphene can be manufactured at an industrial scale, both as powders and dispersions in tonne quantities and as monolayer sheets in large areas.

The International Standards Organisation (ISO) defines graphene as a two-dimensional material up to 10 layers thick, beyond which the electrical properties of the material are not distinct from those for the bulk material, known as graphite [2].

Graphene powders are no longer the only option when considering industrial quantities of graphene. Manufacturing technology and techniques have been evolving rapidly and continuous production processes have been developed that make monolayer graphene with large crystal domains at metre scale and at speeds up to one metre per minute [3].

Multi-layer graphene exists in nature as graphite which consists of a stack of micron sized graphene platelets parallel to the basal graphene plane. Highly oriented pyrolytic graphite (HOPG) is a synthetic form of graphene where the stacks of nanoplates are well aligned with each other with minimal mosaic spread angles which usually occur in natural graphite. While the high temperatures and tensile stress applied in the synthesis of HOPG result in highly ordered structures, there still exist around 0.3° to 0.5° of mosaic spreads and inherent platelet sizes in microns [4].

Figure 1 shows a new form of graphene VdW homostructure. This is a bulk material made of perfectly stacked multi-layers of single crystal graphene. Such a material, when synthesised, would show zero mosaic spreads and large crystals in plane with perfect edge structures. To date, this has not been reported to occur in nature.

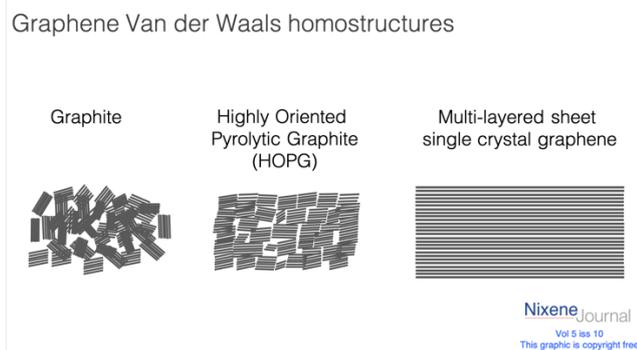


Figure 1: Graphene Van der Waals homostructures

This new VdW homostructure would be a single stack of many hundreds or thousands of layers of graphene in the z dimension and in the x and y dimensions have single crystal domains at centimetre, metre and larger scales. Early indications are that the physical properties of this material will be of considerable academic and commercial interest. For example, the material will be highly rigid, have a very high electrical and thermal conductivity combined with very high tensile strength [5]. The descriptive language of graphite may need further development to describe this material.

This paper will focus on what is known about multi-layered graphene with the intention of stimulating research interest to explore this new area. It begins with monolayer CVD sheet graphene and then moves to multi-layer graphene before exploring the early indications of the physical properties of large-area multi-layer sheet graphene.

2.0 Mono-layer sheet graphene

The term CVD graphene is commonly used to describe large-area sheet graphene (defining “large-area” to be square centimetre and bigger) by a synthetic route. Over the past decade graphene manufacturing has been developing steadily and this material is now routinely made. The Graphene Flagship and Aixtron have built the Neutron roll-to-roll machine capable of making up to 20,000 square metres per year of CVD graphene [6].

2.1 Chemical vapour deposition graphene

Large-area sheet graphene is made by the chemical vapour deposition (CVD) technique. This involves heating a carbon-containing gas or vapour (usually methane) to high temperatures and depositing a layer of graphene on a flat catalytic surface, usually copper

foil although other materials can be used [7]. Once the metallic surface is covered with graphene it can be removed either by peeling or by dissolving the metal.

2.2 Large-area sheet CVD graphene

CVD graphene is produced in research laboratories up to a few square centimetres in size using batch processes.

High quality graphene is also being made industrially up to hundreds of square centimetres in batch processes such as those used by Grolltex Inc in the USA and Graphenea in Spain [8].

General Graphene Corporation (USA), is making monolayer graphene on copper foil with a continuous, roll-to-roll, horizontal CVD process. At the time of writing their generation 3 CVD furnace has the capacity to manufacture 100,000m² per year and will be operational in the last quarter of 2021[9].

Korean company LG Electronics has a roll-to-roll vertical CVD furnace that can manufacture monolayer graphene on copper foil 400mm wide at a speed of 60m/hour (1m/min) [10]. Another Korean company Charmgraphene has announced a roll-to-roll process that can make CVD graphene on 300mm wide copper foil at 120m/hour (2m/min) in sections up to 1km in length [11, 12].

2.3 Polycrystalline and single crystal graphene

A single crystal is defined as consisting of an uninterrupted repetition of the unit cell [13]. A polycrystalline VdW layer consists of a patchwork of individual grains separated from each other via grain boundaries that create domains of different crystalline orientations [14].

Polycrystalline domains are a by-product of current synthesis technology which is an interplay of the two competing forces of growth rate and suppression of nucleation. Greater the number of graphene nuclei, smaller is the size of the crystal before it encounters an incoming growth front and forms a defective boundary. These crystal domains can take the form of hexagons, stars or snowflakes depending on the growth conditions adding to the complexity of controlling size, shape and eventual functional quality which is typical of polycrystalline materials.

Grain boundaries are effectively lines of defects. For example, defects can take the form of carbon rings made from pentagons (5) and heptagons (7), called Stone-Wales defects which are non-periodic. Figure 2 shows a discontinuity combination of 5 and 7 that is called a “5/7 pair” [15].

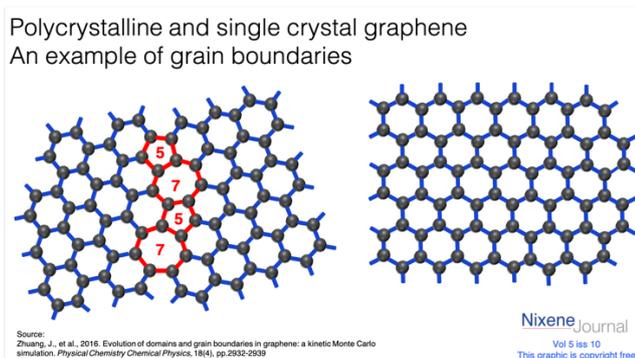


Figure 2: Polycrystalline and single crystal graphene

Defects can also take the form of vacancies where one or more carbon atoms are missing. At the time of writing there is no standard definition of polycrystalline graphene.

The ideal is to form a defect-free sheet of a single grain boundary-free lattice of graphene. This is called single crystal graphene. The longest sheet of single crystal graphene was made at a scale of 500mm by 50mm at Peking University, Beijing, China in 2017 [16]. The largest area of single crystal graphene to-date has been reported by researchers from Oak Ridge National Laboratory, who demonstrated the manufacture of single crystals of 300mm-by-300mm area [17].

2.4 Quality control of CVD graphene

Sun et al., (2021) discussed the industrial scale quality control techniques for optimising CVD graphene production [18]. Raman spectroscopy and terahertz time domain spectroscopy (TTD) are useful for batch processes. Inline roll-to-roll techniques have been more of a challenge. The current view of the authors is that “confocal laser scanning microscopy (CLSM) is expected to satisfy the demands for real-time assessment regardless of production scale. The CLSM image contrast between graphene and copper substrate is found to be inversely proportional to the defect density”. The CLSM technique seems promising for defect detection in a continuous industrial process. However, further work will be needed to distinguish between polycrystalline and single crystal graphene.

3.0 Stacking in VdW graphene homostructures

When the graphene layers are in close contact, approximately 0.34nm [19], VdW forces lock the layers in the stack creating a VdW homostructure.

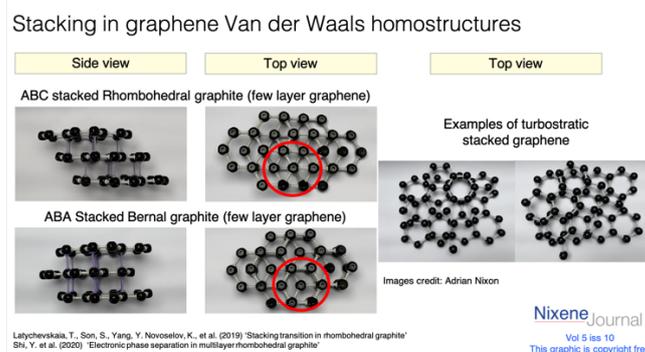


Figure 3: Stacking in graphene Van der Waals homostructures

Stacked layers of graphene can self-assemble in ordered and disordered ways: AB (Bernal), ABC (rhombohedral) and randomly stacked (turbostratic) [20], (Figure 3) depending on how it is formed. The AB form is the most stable and the turbostratic the least stable form [21]. The use of the term VdW homostructure in this paper refers to the AB stacked form unless stated otherwise.

4.0 Multi-layer, large-area sheet graphene

Multi-layer polycrystalline sheet graphene can be made by current methods [22]. The state of the art is to make multi-layer graphene with a CVD process.

Multi-layer graphene can be fabricated in-situ via CVD, or by manufacturing monolayer CVD graphene and artificially stacking individual layers by manual layer transfer process.

4.1 Multi-layer sheet graphene made by metal induced layer exchange methods

Over the years researchers have tended to use nickel as the catalytic substrate to deposit multi-layers. The Ni-induced layer exchange method is one example [23]. This method can produce one hundred to two hundred layers of polycrystalline graphene with an electrical conductivity higher than that of highly ordered pyrolytic graphite (HOPG).

4.2 Multi-layer sheet graphene made by the manual transfer method

Monolayer graphene can be transferred from its host substrate, post CVD, via several techniques which are usually variations of a polymer assisted wet transfer process. In this technique, CVD graphene is supported via a spun-on polymer which acts as a mechanical support while the underlying metal substrate is etched away in an etchant. This leaves the graphene-polymer film floating on the etching liquid which can be transferred to any substrate.

For example, the team at Ulsan National Institute of Science and Technology (UNIST) in Korea made CVD graphene on copper [5], they then added a polymethylmethacrylate (PMMA) layer and dissolved the copper. They floated the graphene on water, then added another layer and repeated this process to create 100 layers of graphene. The team cast camphor onto the last graphene layer of the stack and removed the PMMA with solvent. The camphor supported the stack enabling it to be transferred over a hole in a graphite stage. The camphor was removed by heating to 400°C. This made a freestanding graphene stack 2.5cm² that was then annealed at 2800°C to heal defects and remove intercalated contaminants. Figure 4 illustrates the process.

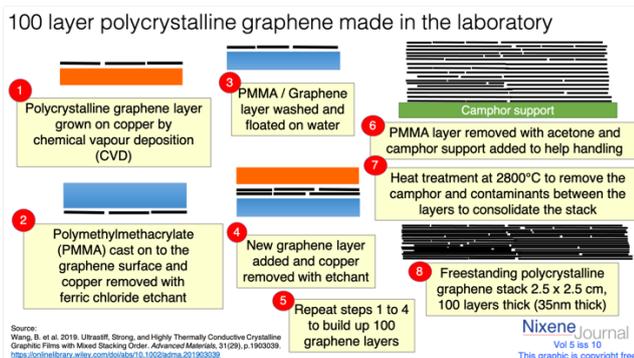


Figure 4: Multi-layer sheet graphene made by wet transfer

The UNIST team examined the freestanding stack at high magnification. High angle annular dark field images showed the freestanding graphene stack contained large variations of contrast that were caused by the presence of both voids and metallic nanoparticles throughout the film. The nanoparticles were copper from the catalytic substrate and iron from the etching solution [5].

So even after the annealing process the freestanding multilayer graphene stack contained voids and

contaminants from the wet transfer process. Before the annealing process the layers of wet transferred graphene would contain adsorbed water and other contaminants that increase the layer spacing and make it less likely that the multilayer graphene would be a VdW homostructure.

General Graphene has created 12cm² samples using a wet transfer process but with no annealing step (Figure 5).

Manufacturing of large-area sheet graphene by a roll to roll process
The first look at test samples of polycrystalline multilayer graphene

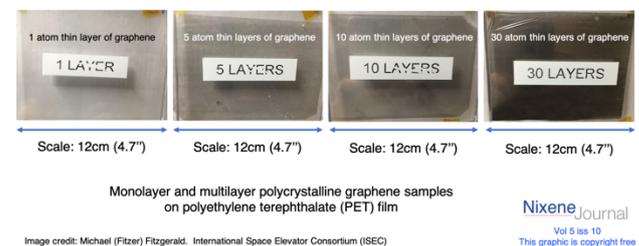


Figure 5: Multi-layer large area sheet polycrystalline graphene made by a wet transfer method

These examples show that multilayer graphene is already being made by industrial scale processes.

5.0 Hypothesis 1: Multi-layered graphene stacks are different to a graphene VdW homostructure

A hypothesis is proposed that non-annealed films of multi-layered graphene prepared via the wet transfer process have graphene layers that are separated by relatively large distances in the stack, by the intercalated contaminants, compared with those post annealing. This implies that the non-annealed graphene stack is not held together as a VdW homostructure because of the larger inter-layer distances. Such a stack, will not benefit from the increase in rigidity that is characteristic of a VdW homostructure.

This hypothesis further suggests that an annealed graphene stack will remove most of the contaminants allowing intimate contact between the stacked graphene layers. This would potentially allow VdW forces to lock together the layers, creating a homostructure. This VdW homostructure would be expected to have increasing rigidity as more layers are added.

6.0 The properties of multi-layer large area sheet polycrystalline sheet graphene

Wang et al., (2019) fabricated 100-layer free standing sheet graphene samples at 2.5cm² scale via the layer transfer method to test its physical properties. The multilayer graphene was reported to have key figures of merit closer in magnitude to a VdW homostructure than a multilayer graphene stack [5]. Figure 6 summarises some of the results.

100 layer VdW homostructure polycrystalline graphene tested in the laboratory

Test	HOPG Graphite [a]	Freestanding 100 layer VdW sheet polycrystalline graphene [a]	Monolayer sheet Single crystal graphene [b]	Multilayer VdW sheet single crystal graphene
Interlayer spacing by X-ray Diffraction	0.336 nm	0.340 nm	Not applicable	Not available
Tensile strength (macroscale)	0.08 GPa	0.7 GPa	130 GPa	Not available
Young's Modulus	20 GPa	62 GPa	1000 GPa	Not available
Thermal conductivity	2000 Wm ⁻¹ K ⁻¹	2292 Wm ⁻¹ K ⁻¹	5000 Wm ⁻¹ K ⁻¹	Not available
Electrical conductivity	0.17 x10 ⁶ S m ⁻¹ [c]	0.22 x10 ⁶ S m ⁻¹	96.0 x10 ⁶ S m ⁻¹	Not available

The freestanding 100 layer sheet polycrystalline CVD graphene was made by the wet transfer method and annealed at 2800°C to remove metal contaminants. The individual layers experienced damage during the transfer process which accounts for the lower values compared with single crystal graphene

Sources:
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Figure 6: Physical properties of 100-layer sheet polycrystalline graphene and graphite

The measured properties for the multi-layer sheet polycrystalline graphene exceeded those for highly oriented pyrolytic graphite (HOPG) [5]. The 100-layer graphene did not match the properties for monolayer single crystal graphene. This is likely to be because the 100-layer graphene is polycrystalline and has multiple vacancy defects caused by the wet transfer process.

7.0 Multi-layer large-area sheet single crystal graphene VdW homostructure

State of the art large-area sheet graphene manufacturing can be summarised as follows:

- Single-layer large-area polycrystalline graphene. [6, 8, 9, 10, 11]
- Multi-layer large-area polycrystalline graphene VdW homostructure. [5]
- Single-layer large-area single crystal graphene. [16]

As of the time of writing of this paper, multi-layer large-area single crystal graphene VdW homostructure has not yet been reported and therefore presents an unexplored niche in the field of graphene and layered materials research.

8.0 Hypothesis 2: A VdW homostructure of large-area sheet single crystal graphene represents research territory that is yet to be explored

At the time of writing a VdW homostructure of multi-layer, large-area single crystal graphene has not been reported. The hypothesis is that this new material has a combination of fascinating properties that will provide a fruitful area of research for the future.

8.1 Some physical properties of VdW graphene homostructures

The following examples have been selected to illustrate the probable characteristics of this new advanced material.

8.1.1 Strength

There are mixed messages in the literature about the strength of few-layer graphene.

Falin et al., (2017) compared few-layer graphene and few-layer hexagonal boron nitride (hBN) [24]. The mechanical properties of these materials were studied by indentation at the centre of regions suspended over a micro-well fabricated in and SiO₂/Si substrate using atomic force microscopy (AFM). The breaking strengths of graphene and hBN nanosheets of different thicknesses were calculated based on load-displacement curves and fracture loads using finite element simulation based on the deflection of the AFM tip. The work found that the breaking strength of graphene decreased by more than 30% (while still being a comparatively very strong material) when the number of layers increases from 1 to 8, while the breaking strength of hBN nanosheets was not sensitive to increasing thickness.

The team reported that when strain and compression was applied, such as that experienced by the region close to the AFM indentation centre, graphene and hBN exhibit different layer sliding energies. The sliding energy in graphene tends to zero and in hBN the sliding energy increases. This means that under compression graphene layers slide over one another while hBN layers resist sliding.

In this strength test, the team realised that sliding could concentrate stresses in the lowest layer bonded strongly to the SiO₂ substrate. This effectively shielded the other layers. Extra layers added to graphene did not proportionately add to the load bearing capacity, whereas in hBN additional layers make a proportional contribution to the load bearing capacity. This explains why the AFM test produced lower breaking strength values for multilayer graphene and higher breaking strength values for multi-layer hBN in the localised region of indentation.

Mazilova, et al., (2018) used a field ion microscope to determine the uniaxial tensile strength of graphene nanosheets at low temperatures [25]. The work showed that for multi-layer graphene 0.5 to 2.8 nm thick, the maximum strength within the measurement error does not depend on the thickness of the sheets.

8.1.2 Strength and lightweighting

A VdW homostructure of single crystal graphene will be very strong and lightweight. A material with these properties will find many applications.

One application is the material to make the tether for a space elevator. A space elevator can be thought of as a vertical railroad into space. A tether (cable) stretches from the ground to an Apex Anchor (counterweight) 100,000 km up/out in space. Elevator cars (tether climbers), travel up and down the tether and carry cargo and eventually humans to and from space. The engagement of the drive mechanism to the tether is a critical consideration and is called the climber-tether interface. The International Space Elevator Consortium (ISEC) has an active study group of scientists and engineers researching the climber-tether interface [26].

The tether material has to support its own weight as well as that of the climber. This means the material has to be very strong and lightweight. ISEC calculates that a tether made from 12,333 layers of single crystal graphene, less than 1 metre wide and 8 microns thick will hold a 20-tonne climber with a 40% safety margin. Each square metre of this layered tether material will weigh just 9.5 grammes [27].

Wright, et al., (2021) reports that the ISEC study group is starting to converge on the view that a tether made from a VdW homostructure of multi-layer single crystal graphene will probably be climbable [26]. This means a space elevator climber can be designed and built

from existing materials to grip the tether and be capable of pulling itself up a tether made from a VdW homostructure of single crystal graphene.

8.1.3 Rigidity and stiffness

For the purposes of this paper, we will assume that rigidity and stiffness are interchangeable terms. Rigidity is an important factor to consider when designing structures such as the space elevator tether [28]. Work done on graphene at the micro scale has found that the rigidity of multi-layer graphene increases with the cube of the number of layers [29].

Rigidity of multilayer sheet single crystal graphene scales with the cube of number of layers in a VdW homostructure

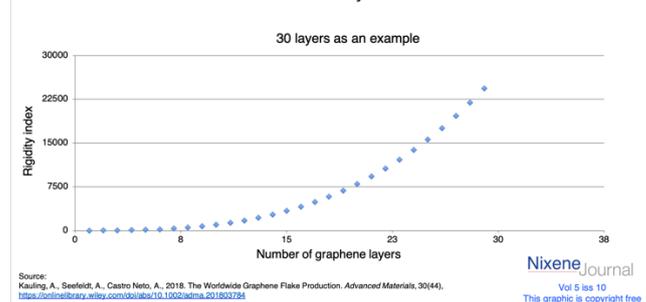


Figure 7: Rigidity of multi-layer graphene scales with the cube of the number of layers in a VdW homostructure

Recent work at the King Abdulaziz University and University of Adelaide found that the bending stiffness of multi-layer graphene scales with the square of the number of layers [30, 31]. However, this only applies to seven layers or less of graphene.

Wang et al., (2019) also found that, when layered, polycrystalline graphene created an ultra-stiff bulk material [5]. The experimental findings confirm the escalating rigidity or stiffness of the multi-layered material.

On extrapolation of this study, it can be envisaged that as sheets of single crystal graphene are layered to create a VdW homostructure, the bulk material becomes increasingly rigid. This is an important property to understand for very large-area applications of the future such as the tether for a space elevator which would need over 12,000 layers of single crystal graphene.

8.1.4 Thermal properties

Existing multi-layer graphene has anisotropic thermal conductivity. This means the physical properties are very different in different dimensional planes. The in plane thermal conductivity (x-y direction) is at least $2000 \text{ Wm}^{-1}\text{K}^{-1}$ and the cross plane thermal conductivity (z direction) is several hundred times lower at $6.8 \text{ Wm}^{-1}\text{K}^{-1}$ [32]. A VdW homostructure of single crystal multi-layer graphene will enhance these properties.

Graphene has an extremely high melting point. The latest estimates place this somewhere between 4000K and 6000K for a freestanding monolayer of graphene [33]. Savvatimskiy et al., (2017) have studied the thermophysical properties of HOPG and found the melting point to be up to 5000K [34]. This indicates the melting point of a VdW homostructure of single crystal graphene will be of a similar order of magnitude.

Experiments and molecular dynamics simulations reveal that multi-layer graphene resists burning in oxygen. The more single-crystal-like the graphene the more resistant to burning is the bulk material even up to 2000K in oxygen [35]. This is because burning of graphene layers starts at defect sites. Multi-layered single crystal graphene has fewer defect sites than polycrystalline graphene containing defects and so resists burning to a much greater extent.

VdW homostructures of multi-layer large-area sheet single crystal graphene would therefore find many applications in thermal management structures such as hypersonic applications [36].

8.1.5 Gas impermeability

The University of Manchester has explored the gas permeability of single crystal monolayer graphene. A one atom thin layer of graphene is as impermeable to helium gas as a kilometre thick wall of glass [37].

Hydrogen gas can dissociate into protons that will pass through monolayer single crystal graphene. However, bilayer graphene is impermeable to hydrogen [38].

Hydrogen is one of the main candidates to replace fossil fuels [39]. However, storing and transporting the gas is a challenge. Hydrogen can leak through many materials and can also cause embrittlement of metals leading to the loss of integrity of the containment [40].

Work has yet to be done exploring the hydrogen permeability of multi-layer large-area polycrystalline graphene VdW homostructures.

Future applied research could be directed towards using VdW homostructure multi-layer large-area single crystal graphene to create hydrogen gas impermeable coatings or containment structures because these will be needed to realise any forthcoming hydrogen economy.

8.1.6 Optical characteristics

An interesting question to ask is 'what does multi-layer large-area sheet single crystal graphene look like?'

Early demonstrations of multi-layer sheet polycrystalline graphene look very dark grey or black as shown in Figure 5. When describing the appearance of this material, a common response from researchers is to say it will be black, or very dark grey. Most people are used to thinking about multi-layer graphene in terms of graphite and graphite powders. Detailed work on the reflectance of multi-layer sheet graphene is only recently being explored.

One study from 2016 [41], measured the reflectance of one, two and three layer graphene and found that as the layers increased the reflectance decreased with a pronounced effect at wavelengths around 600nm as shown in Figure 8.

Normalised reflectance of 1,2 and 3 layer graphene as a function of wavelength between 550 and 700nm

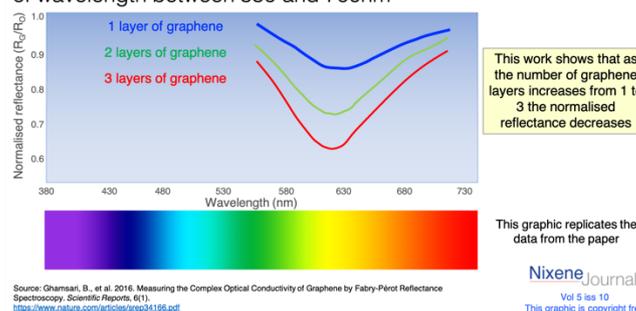


Figure 8: The normalised reflectance of 1,2 and 3-layer graphene

More recent work done at the University of Manchester created 150-layer polycrystalline graphene by the Ni-induced layer exchange method [42]. The work investigated the reflectance spectrum of the multi-layer polycrystalline graphene and found a broad reflectance of approximately 20% across the visible spectrum as shown in figure 9. This indicates that the multi-layer graphene would have a perceived metallic colourless/silvery appearance because all wavelengths of light in the visible part of the spectrum are reflected equally.

The team also discovered that this multi-layer graphene has very high reflectance in the terahertz (THz) and microwave regions of the spectrum.

Reflectance of 150 layer CVD graphene encased in polyethylene

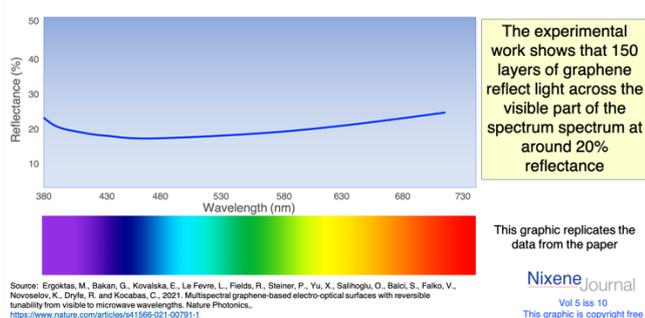


Figure 9: The reflectance spectrum of 150-layer polycrystalline graphene

The work also explored the effect of charge states on the optical properties of the multi-layer graphene and, to achieve this, the multi-layer graphene was intercalated with lithium. The presence of intercalated species between the layers seems to have created a darker appearance than the multi-layer graphene samples.

This intercalation could explain why the samples in figure 3 appear increasingly dark with increasing layers. More work is required to explore this, perhaps by high temperature annealing of the wet transferred graphene, as done by the team at UNIST.

Prof. Coskun Kocabas presented another sample of multi-layer, large-area, polycrystalline sheet graphene at a University of Manchester, Graphene Engineering Innovation Centre (GEIC) webinar [43].

Chemical vapour deposition (CVD) multi-layer large-area sheet polycrystalline graphene transferred to a paper substrate

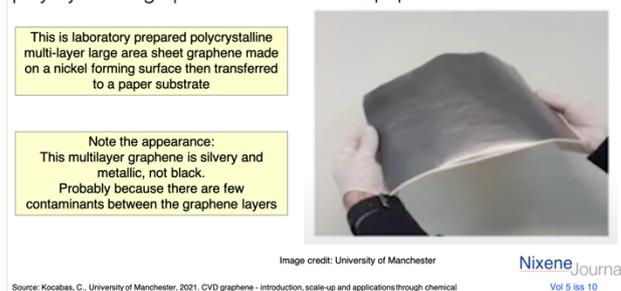


Figure 10: multi-layer large-area polycrystalline graphene

The multi-layer graphene was made in the laboratory on nickel then transferred to a paper substrate as a laboratory proof of concept. Figure 10 shows the appearance of this material.

It can therefore be envisaged that a multi layered large-area single crystal graphene VdW homostructure would exhibit specular reflectance where normal graphite exhibits diffuse reflectance as illustrated in figure 11 [44].

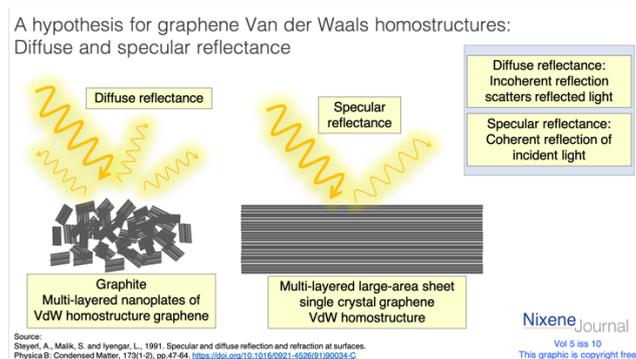


Figure 11: Hypothesis for diffuse and specular reflection in graphene VdW homostructures

Experimental realisation of such a material therefore could open up the possibility of creating very lightweight, rigid and thermally stable broad-spectrum mirrors that could find use in space applications.

The suggestion is that the range of exceptional properties that monolayer graphene possesses may carry over, in whole or in part, to the bulk layered material as a VdW homostructure. This is why the term frontier material has been applied in this case.

Researchers are already starting to explore possible routes to synthesise and demonstrate these hypotheses. For example, Prof. Rodney Ruoff's team at the IBS Center for Multidimensional Carbon Materials located at UNIST in South Korea has indicated a developing interest in what they are calling graphitic films [45].

9.0 Summary

Polycrystalline graphene is now manufactured by industrial processes at speed and scale. It is starting to be made in tens and hundreds of layers. This layered material is currently considered as graphite and therefore is considered to be of lesser interest and value. The same industrial companies also have the capability to make single crystal graphene.

This paper highlights multi-layer, large-area sheet single crystal sheet graphene as a VdW homostructure with a layer spacing of 0.34nm. Far from being dismissed as 'just graphite', this should be considered a new advanced material. Hypotheses are proposed that suggest this material may have multiple superlative physical properties which would enable applications such as ultra-light broad-spectrum mirrors for space applications and when manufactured at scale might possess the mechanical properties that could eventually enable the long-awaited space elevator tether that would make future space travel energy and resource efficient.

The term multi-layer, large-area single crystal sheet graphene Van der Waals homostructure, is rather cumbersome and so in the absence of a better alternative a new descriptive term is required.

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Affiliations

Adrian Nixon is a member of the board of directors of Nixene Publishing Ltd and Editor of the Nixene Journal. He is a member of the board of directors of the International Space Elevator Consortium and an advisory board member for Stellar Modal Space Transportation. Ravi Sundaram is head of strategic R&D markets at Oxford Instruments UK and board advisor at Nixene Publishing Ltd. Chris Bentley is a director at the Strategy Exchange Ltd and board advisor at Nixene Publishing Ltd.

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Correspondence and requests for materials should be addressed to Adrian Nixon at Nixene Publishing using the email address info@nixenepublishing.com. This revised version contains corrected values for tensile strength and Young's modulus of 100 layer polycrystalline graphene in table 6. Also, a revised first paragraph that refers more precisely to graphene isolation.

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