Things you could do with graphene

Graphene and its derivatives are very versatile materials. As a result of their electrical, mechanical and optical properties they have, in the past 10 years, been proposed for use in a wide range of applications ranging from flexible electronics to DNA sequencing. They offer the promise to significantly improve existing products and to enable the design of materials and devices with novel functionalities. We asked seven experts in their fields to share their views on the potential and challenges in the realization of graphene-based products in a range of technologies. Development of graphene materials and devices is more mature in some areas, such as flexible electronics and conductive inks, but is at a more conceptual stage in others, such as biomedical and aerospace technologies. Several challenges towards commercialization remain that are application-dependent. Furthermore, a common problem for the introduction of graphene in industrial applications is the lack of methods for the large-scale production of graphene materials, in particular of electronic-quality layers.

Graphene for displays that bend

Jong-Hyun Ahn and **Byung Hee Hong** discuss how graphene can be used in the development of flexible electronics.

Electronic components that are flexible and stretchable are more versatile than rigid ones, and are sought after for optoelectronic devices such as displays, solar cells and light emitters. Flexible components need to be fabricated on soft substrates such as plastic or rubber, but traditional fabrication methods use a rigid substrate, and therefore new materials and processes are required. In optoelectronic devices, transparent electrodes are key components, and at present are typically made from indium tin oxide (ITO) because of its relatively good conductivity and optical transparency. But ITO has poor mechanical properties: when bent or stretched it tends to crack, and this leads to a deterioration of its electrical properties.

Graphene has emerged as a promising material for transparent and flexible electrodes. The optical transmittance of monolayer or few-layer graphene grown by chemical vapour deposition (CVD)[1] is superior to that of ITO and other emerging materials, and the sheet resistance of graphene films is comparable to that of ITO because of graphene's high carrier mobility ($>10^3$ cm^2 V^{-1} s^{-1}) and high carrier density ($>10^{12}$ cm^{-2} for doped samples)[2]. Furthermore, graphene on flexible substrates is mechanically robust, and has the smallest bending radius among all flexible

transparent conductors characterized so far[3]. Synthesis by CVD is also advantageous because it consumes only tiny amounts of carbon gases and does not need rare mineral resources such as indium or silver. The catalytic metal substrate is also reusable, and recycling technology is industrially available.

The use of graphene as a transparent electrode has already been demonstrated in a variety of flexible optoelectronic devices, including touch-screen sensors[4] (Fig. 1), organic light-emitting diodes[5] and organic photovoltaic devices[6]. The

possibility of fabricating lightweight, thin and low-cost flexible electronics devices through continuous roll-to-roll processes is another important advantage of using graphene electrodes[4].

Despite this, considerable challenges must be overcome to integrate graphene-based transparent electrodes into commercial devices. These include the development of a low-cost, large-scale synthesis method for high-quality graphene with guaranteed uniformity and reproducibility[7]; a defect- and residue-free transfer method that
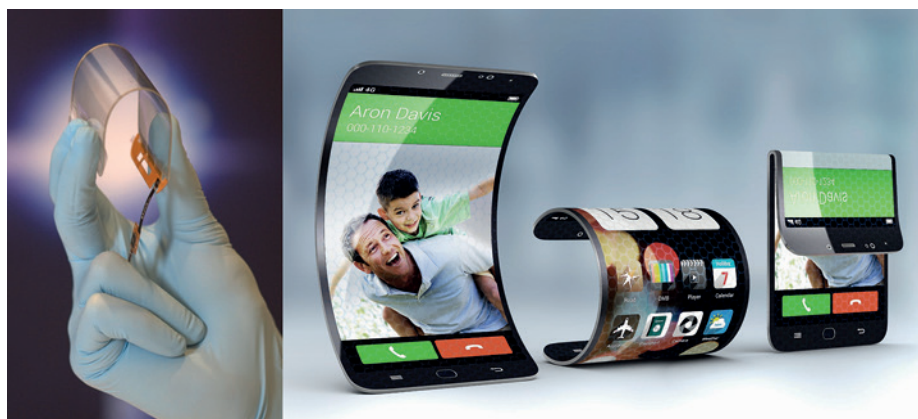

Figure 1 | Graphene-based multi-touch screen showing excellent flexibility (left)[7] and possible applications in bendable or foldable mobile devices (right).

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is compatible with conventional device manufacturing processes[8]; doping processes that can assure stable, high electrical conductivity over long periods[9]; a method to improve the environmental stability of graphene electrodes against moisture and chemicals in the air[10]; and a method to decrease the contact resistance between electrodes and active materials[11].

One issue, in particular, with CVD-grown graphene is that it must be transferred to a perfectly flat surface. Most electronic devices, however, have an interconnected, multilayered structure. Although this is not a problem for ITO, as it can be sputtered onto irregularly shaped surfaces, the device architecture needs to be customized for two dimensions when using graphene electrodes. This limits the immediate replacement of ITO by graphene. Because of this, we anticipate that applications to flat and simple structures

such as touch screens, smart windows, electromagnetic interference shields, lighting and transparent heaters[12] will be the first to be realized, whereas applications to flexible displays and microelectronic devices will follow some years later.

We expect that the market for flexible electronics will become larger than that for non-flexible electronics in about 10 years. Although there are many candidates for ITO replacement — including metal meshes and silver nanowires whose sheet resistances are as low as ~10 Ω sq^{-1} at ~90% transmittance[2] — none of them are as flexible as graphene. For example, the conducting properties of graphene can remain stable up to ~11% extension[1], and its initial sheet resistance persists for ~300,000 cycles at 3% bending strain[7], whereas a silver nanowire breaks easily at strains of less than 1%[13].

Resolving the above-mentioned challenges will bring closer the advent of

flexible electronics, in which graphene technology will be as stretchable as our imagination. ❑

Jong-Hyun Ahn is at the School of Electrical and Electronic Engineering, Yonsei University, Seoul, Korea. Byung Hee Hong is in the Department of Chemistry, Seoul National University, Seoul, Korea.
e-mail: ahnj@yonsei.ac.kr; byunghee@snu.ac.kr

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Electrifying inks with 2D materials

Felice Torrisi and **Jonathan N. Coleman** describe how graphene can be used in conductive inks to print electronic circuits.

Printed electronics[1,2] has recently emerged as an important new direction for electronics beyond conventional silicon-based technologies. Electronic devices, including radiofrequency identification tags, light-emitting diodes, batteries and transistors, can be built by printing liquid-dispersed functional materials such as organic polymers, nanoparticles and nanotubes. Techniques including inkjet, roll-to-roll and spray deposition can pattern these materials on either rigid or flexible substrates, at low temperature and over large areas, and most importantly at very low cost. However, the performance of printed devices is limited by the quality of the materials contained in the inks and how these are arranged on the target substrate. Graphene and other two-dimensional (2D) materials are solution-processable and have useful physical and chemical properties. Can they make an impact on printed electronics?

Electronic inks are mixtures of a functional material, stabilizers and rheology modifiers. Different inks are needed to print the different elements of devices: semiconducting inks (the functional materials) in the active layer, insulating materials for dielectrics, and conducting materials for electrodes. They must be stable, cheap and print easily on appropriate substrates to produce devices with high charge-carrier mobility and long lifetime, without the need for aggressive post-treatments. Finding materials that fulfil these requirements is difficult. For example, conducting inks often contain metallic nanoparticles that may require sintering at high temperatures to maximize conductivity, which is problematic for polymeric

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substrates. Furthermore, polymer-based inks often lack stability whereas nanotube inks are generally unsuitable for semiconducting applications owing to the presence of both metallic and semiconducting nanotubes.

Graphene has the potential to fulfil most requirements for use in conducting inks thanks to its high carrier mobility, mechanical robustness, environmental stability and potential for low-cost production. Before an ink can be produced, graphene must first be dispersed as nanosheets in a liquid. Graphene suspensions can be produced from graphite by oxidation to produce graphene oxide, or by liquid-phase exfoliation in solvents to give few-layer nanosheets[3]. Without high-temperature or aggressive chemical processing, however, it can be hard to achieve high conductivities in films produced from graphene oxide nanosheets. On the other hand, large quantities of pristine nanosheets, typically hundreds of nanometres across and ~1 nm thick, can be produced quickly and easily by liquid-phase exfoliation in readily printable liquids such as water and organic solvents[4]. The resulting ink is stable, processable in ambient conditions, and has high batch-to-batch reproducibility as well as good rheological properties for printing and coating[5]. Continuous networks of graphene nanosheets have been produced by methods such as spraying, filtration and rod coating, and tend to have conductivities of up to 10^4 S m^{-1} (ref. 6), even without high-temperature or chemical treatments[7–9]. They have performed well in applications such as electrodes for supercapacitors, as well as transparent electrodes for liquid-crystal displays, thin-film transistors and smart windows[5,8,9]. Patterned networks, with features as small as 30 μm (ref. 10), have been produced by inkjet printing[5,7] and gravure coating[10], and have shown promise in applications from supercapacitors[11] to antennae[12].

Although graphene can be used in flexible interconnects or as an active material in printed supercapacitors or composite devices[5,9], its lack of a bandgap means that it cannot replace conjugated polymers[2] as the semiconducting material in printed electronic devices. In addition to graphene, however, liquid-phase exfoliation can produce suspensions of a range of other 2D nanosheets, including WS_2, MoO_3 and BN (ref. 3). These 2D materials have diverse properties, with some being semiconductors or insulators and some electrochemically active, making their inks suitable for a wide range of applications as active materials in printed electronics. Spray coating[13] or inkjet printing[14,15] of MoS_2 nanosheets have been recently reported for supercapacitor and sensing applications.

But the most exciting applications of 2D materials in printed electronics will come from printing devices where two or more nanomaterials are integrated together in well-defined structures. All-inkjet-printed heterostructure photodetectors and transistors have already been produced by sequential printing of graphene and WS_2, MoS_2 or BN (refs 7,16). It is likely that many developments will stem from these pioneering works.

Nevertheless, a number of important hurdles remain before these developments can lead to commercialization of 2D material-based inks. For example, although nanosheets can be made in large quantities by liquid-phase exfoliation, the yield of monolayers is is still low, typically on the order of 5–25%, and the flake size is poorly controlled. For inkjet printing, the nanosheet size and the rheological properties of the suspension must be carefully tuned[5]. Furthermore, the high aspect ratio of nanosheets generally results in low-concentration suspensions (~0.01–1 wt%), which is problematic for some printing techniques. Electrical performance is another issue. Printed devices will consist of networks of either graphene or inorganic nanosheets, and charge transport will be limited by junctions between the sheets, resulting in mobilities typically two orders of magnitude lower than in individual nanosheets[17]. This will limit the achievable conductivity of graphene interconnects, and reduce the performance of devices based on semiconducting nanosheets. On the other hand, as MoS_2 nanosheets have mobilities of hundreds of cm^2 V^{-1} s^{-1}, production of MoS_2 networks with mobilities of a few cm^2 V^{-1} s^{-1} might be possible, making them competitive with printed conjugated-polymer-based devices[2].

We believe that the future is bright for printed devices based on 2D materials, although many challenges remain. As new materials combinations are demonstrated, the range of printed devices and applications will expand. Moreover, as the scaling up of ink production proceeds, costs will fall, eventually allowing the best 2D material-printed devices to compete with traditional technologies on cost and possibly performance. ❐

Felice Torrisi is at the Cambridge Graphene Centre, Engineering Department, University of Cambridge, 9 JJ Thomson Avenue, Cambridge CB3 0FA, UK. Jonathan N. Coleman is at the CRANN and AMBER Research Centres and the School of Physics, Trinity College Dublin, Dublin 2, Ireland. e-mail: colemaj@tcd.ie

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Charging graphene for energy

Energy storage is a grand challenge for future energy infrastructure, transportation and consumer electronics. **Jun Liu** discusses how graphene may — or may not — be used to improve various electrochemical energy storage devices.

Graphene has captured the imagination of researchers for energy storage because of its extremely high theoretical surface area (2,630 m^2 g^{-1}) compared with traditional activated carbon (typically below 1,500 m^2 g^{-1}), excellent electrical conductivity, high mechanical strength and potential for low-cost manufacturing. These properties are very desirable for achieving high capacity and energy density, and fast charge and discharge rates. Graphene sheets used in energy

storage research are usually prepared by oxidation and reduction of graphite[1]. Even though these chemically derived graphene materials contain multiple stacked graphene sheets, structural defects (such as vacancies) and oxidized functional groups, they are more suitable for energy storage applications than single-layer graphene, because they can be made in relatively large quantities. In the literature they are also called reduced graphene oxide, or functionalized graphene sheets, but here they are all referred to as graphene for ease of discussion.

Two important devices — batteries and electrochemical capacitors — have been widely investigated. In a battery material, the redox reaction occurs at a stable voltage, and the energy is stored in the bulk. Therefore, the energy density is high (more than 100 W h kg^{-1}), but it is difficult to charge or discharge the device rapidly (power density is less than 1 kW kg^{-1}; ref. 2). In an electrochemical capacitor (also called supercapacitors or ultracapacitors in the literature), the energy is stored as adsorbed ionic species at the interface between the carbon surface and the electrolyte. Thus, charge and discharge can be rapid (power density up to 10 kW kg^{-1}) but the energy density is low (less than 10 W h kg^{-1}). A device that could posses both high energy and high power would be ideal.

Graphene is a great fit for electrochemical capacitors. The fact that ion adsorption occurs at the surfaces, and on the edges, of the sheets means that the potential at which the process takes place varies continuously as a function of the surface charge coverage, a primary requirement for supercapacitive materials. Furthermore, the unique combination of high surface area and excellent electrical conductivity makes graphene even more attractive[3]. The theoretical specific capacitance of graphene can be as high as 550 F g^{-1} when all the surface is used. As a comparison, typical commercial electrochemical capacitors using activated carbon have a capacitance of about 100 F g^{-1}. At present, the theoretical capacity has not been achieved because graphene sheets tend to restack in the electrode, but values in the range 200–300 F g^{-1} have been reported[4].

Alongside electrochemical capacitors, graphene has garnered enormous interest as cathode and anode material for Li-ion batteries[5]. Graphene sheets can have a specific capacity greater than 500 mA h g^{-1}, which compares favourably with graphite (372 mA h g^{-1}), the anode material used in commercial Li-ion batteries (Fig. 1). For a battery to work, however, the cathode and the anode need to be charged and discharged at different potentials, and the operating voltage window is determined by the difference between the discharge potential of the cathode and the anode. To achieve high capacity, graphene would need to be charged at more than 3 V. Unfortunately, this does not seem realistic, as anode materials should be charged and discharged at less than 1 V. At this potential, graphene actually has a lower specific capacity than commercial graphite. Functionalized graphene sheets have also been studied as cathode material[6], but they mostly exist in a delithiated state. As a result, a Li-rich anode material, such as Li metal, needs to be used. But the use of a Li-metal anode introduces entirely new challenges, as discussed later. Very recently, an all-graphene battery was explored[7], partially reduced graphene sheets are used as the cathode, which can be charged to a high voltage, and graphene oxide is used as the anode. The battery shows both high energy density (225 W h kg^{-1}) and high power density (6,450 W h kg^{-1}), but how to make a practical device remains unclear.

There are other promising ways to exploit the properties of graphene for battery applications. One of them consists in using graphene as a coating material to enable the direct use of Li-metal anodes. Lithium metal has the highest capacity among all the anode materials (3,860 mA h g^{-1}), but it cannot be used as is because it forms dendrites that can potentially cause internal shortage and thermal run-away (fire). In a new electrode design, Li metal was coated with graphene sheets that essentially function as high-surface-area capacitor materials, reducing the current sustained by the Li metal itself, and therefore reducing dendrite growth[8].

Additionally, graphene could be exploited to make composite materials with enhanced electrical conductivity and mechanical properties. It is well documented, for example, that adding graphene to low-conductivity materials such as TiO_2, $LiFePO_4$ and organic materials can aid charge transfer and promote high-rate charge–discharge cycles. Moreover, ordered graphene–SnO_2 sandwich structures or crumpled graphene–Si composites possess enhanced mechanical stability that can accommodate the large volume change experienced by the electrode during lithiation and delithiation for improved cycling stability[9,10]. The porous structure of graphene-based materials, with controlled functionalization density, has also been explored for sulphur encapsulation for Li–S batteries[11,12], devices with substantially higher theoretical energy density (2,600 W h kg^{-1}) than Li-ion batteries[13]. These designs provide new prospects for energy storage, but the research is still in the early stages.
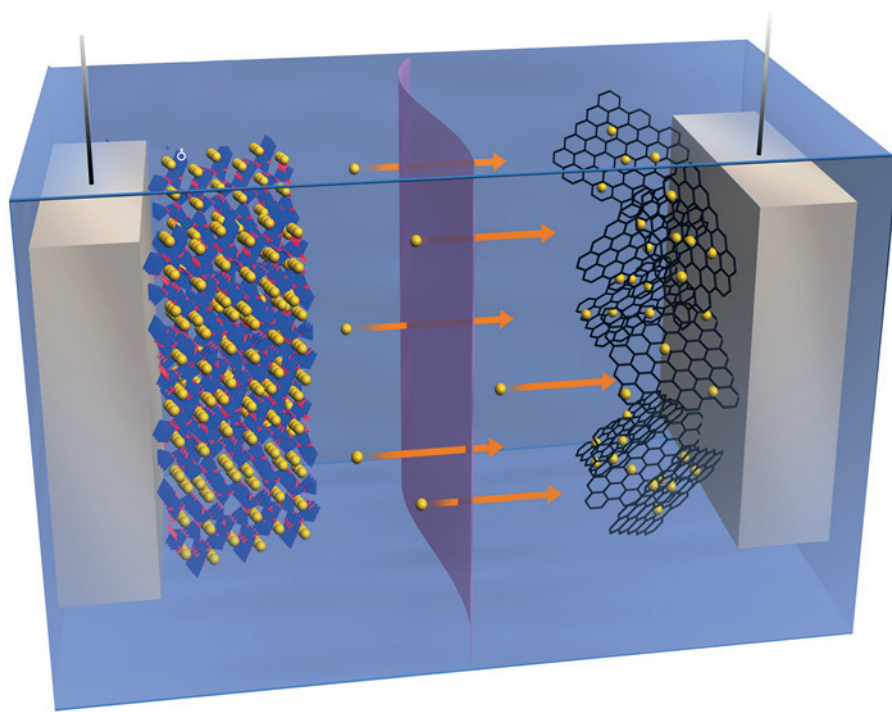

Figure 1 | A schematic representation of a hybrid battery–capacitor device, in which an intercalation material would be used as the cathode (left) and graphene as the anode material (right). Lithium ions (yellow) are inserted and de-inserted during charge and discharge cycles. Figure reproduced with permission from ref. 16, © 2011 Wiley.

An under-explored territory is to use graphene as a platform to understand complex electrochemical reactions, as the surface activity of graphene can be controlled relatively easily by chemical functionalization. This direction could be especially useful to understand Li–air batteries. These devices have the highest theoretical energy density (12,000 W h kg⁻¹) among known battery systems, but the reactions occurring at the carbon cathode to form the product, Li_2O_2, are poorly understood and mostly irreversible[14]. In some initial studies, functionalized graphene sheets have been used to identify the active sites on carbon — mostly defects — where the desired Li_2O_2 phase forms[15].

For practical applications, low-cost and large-scale production methods need to be developed. At present, industrial facilities can produce around 100 tonnes of graphene per year, but the quality and reliability of this material (uniformity, particle sizes, purity, surface area and carbon to oxygen ratio) remain an issue. Moreover, graphene production is still 5 to 10 times as expensive as activated carbon or graphite, commonly used materials in commercial electrochemical capacitors and batteries. For capacitor and battery applications graphene also needs to be made in proper engineered forms. Normally, graphene has very low packing density (as low as 0.015 g cm⁻³), which translates into very low volumetric energy density in a device. In comparison, typical graphite anode materials have a packing density of about 1 g cm⁻³. Graphene sheets also have a tendency to restack, reducing the effective surface area, and may form two-dimensional structures (such as layers parallel to the electrode surface) that hinder ion and electron transport across the electrode. As a result, it is challenging to make practical electrode materials with sufficient thickness (more than 20 μm) to obtain meaningful capacitance or capacity per unit area. Either the materials supplier or the device manufacturer needs to find ways to pack graphene sheets into high-density aggregates without sacrificing the surface area.

It is clear that graphene has great potential, but it is not the panacea for all problems in energy storage. The first real application is likely to come from electrochemical capacitors, assuming that low-cost and high-quality graphene will become available in large quantities and can be made into the desired aggregate structure. Graphene capacitors will be attractive for grid applications that require fast response time (milliseconds), high efficiency (over 95%), high power, long cycling life (more than thousands of cycles), and variation in the frequency caused by the power mismatch of supply and demand. Graphene capacitors will also find applications in capacitor–battery hybrid devices for hybrid and all-electrical vehicles to provide the power for start-up and acceleration.

But claims in popular news that graphene capacitors will make batteries obsolete are premature. The truth is that batteries and capacitors still fulfil quite different functions. In the near future, it is also unlikely that graphene will directly replace some of the widely used anode or cathode materials in Li-ion batteries just because it has a higher specific capacity. To develop practical electrode materials and devices, one must consider the interplay of the intrinsic properties of graphene, the overall structure of the electrode, and the desired properties and functions of the application, as well as how the device will work. In the literature, there is too much focus on achieving high specific capacitance or high specific capacity of graphene without considering whether the whole charge–discharge curve is appropriate for a real device and even whether the positive and negative electrodes will work together. Finally, there is very limited research on how to make the electrode material with a morphology that can deliver good packing density, and how the electrode material should be incorporated into a real, stable device. It is likely, however, that, in the medium term, graphene will be used as an additive or coating material for electrodes in Li-ion and other battery applications to improve the stability, capacity at fast discharge rate and other electrochemical properties. ❐

Jun Liu is at the Pacific Northwest National Laboratory, Richland, Washington 99352, USA. e-mail: Jun.Liu@pnnl.gov

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Graphene against corrosion

Siva Böhm discusses how graphene can be used to prevent corrosion of metals such as steel.

Corrosion is one of the greatest challenges faced by the steel industry. It is triggered by environmental factors, such as water, oxygen and electrolytes. The annual direct cost of metallic corrosion for infrastructure (highway bridges, pipelines, material storage), utilities (drinking water, gas/electric), automotive transportation and government defence is estimated to be US$300 billion in the United States and €200 billion in Europe. Therefore, robust and long-lasting solutions are required to overcome corrosion issues. Until recently, corrosion protection for steel was provided by zinc- and chromium-based coatings; however, because of the adverse effects of chromium on human health and the environment, its supply and use in Europe are restricted. There is an increasing need for more effective, environmentally friendly and less costly products. Furthermore, innovative technologies are shifting the focus from protection against corrosion to its prevention.

Graphene has attracted attention for applications in many industries since its isolation in 2004[1]. A graphene layer is made of carbon atoms arranged in a hexagonal lattice, with each atom bonded to three nearest neighbours within the plane by sp^2 hybridized orbitals. This results in a high inherent strength of 0.142 N m along carbon bonds, which makes graphene (in the absence of defects) the strongest material ever discovered, with an ultimate tensile strength of 130 GPa. Graphene has other attractive properties, including electron mobility up to 2.5×10^5 cm² V⁻¹ s⁻¹, thermal conductivity of more than 3,000 W m K⁻¹, Young's modulus of 1 TPa and the capability

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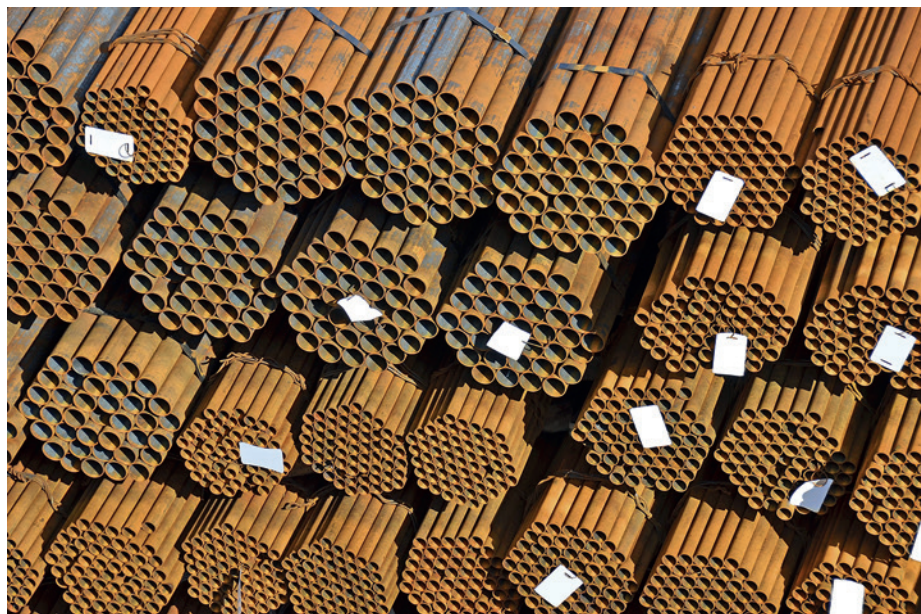
of sustaining high current densities[2]. In particular, impermeability to all gases and salts makes graphene an excellent candidate in anticorrosion coatings.

In the past, fullerenes and exfoliated or intercalated clay materials have been used for anticorrosion applications, but those materials are highly insulating and also hydroscopic. Hydroscopic material acts to suppress the anticorrosion behaviour of the coating by increasing water uptake, weakening the coating's barrier properties. For coated metal substrates to be used in some applications such as fuel cells and solar cells, the coating needs to be electrically conductive, as a fully insulating coating is unsuitable.

Graphene's excellent electrical conductivity and high surface area play an important role in its anticorrosion mechanism. As we currently understand it, the anticorrosion mechanism provided by graphene can be explained by a combination of three processes. On the one hand, graphene coatings can make the path of permeating water more tortuous. Second, because of the impermeability of pristine graphene, the coatings act as an excellent barrier to water, oxygen and other corrosive materials. Furthermore, graphene has a higher electrical conductivity than steel. With polymeric anticorrosion coatings, when the corrosion starts at the metal/coating interface, electrons generated by the anodic reaction move through the metal to a cathodic site, and complete the corrosion reaction. This is the reason that, once the corrosion starts at the interface of the metal and a conventional insulating coating, it is difficult to stop. However, when functionalized graphene is incorporated in

the coating, because of its excellent electrical conductivity, an alternative path for the electrons is provided, so that they may never reach a cathodic site. Moroever, graphene and graphene hybrid nanocomposites can be used as inhibitors in protective coatings to prevent oxidation of underlying metals in oxidizing chemicals, water or air for longer periods[3–7].

Recently, a highly versatile top-down approach was proposed for industrial production of graphene from graphite by liquid exfoliation, which functionalizes graphene and seamlessly incorporates it into organic coatings[3,4,8,9]. Graphene-based coatings showed an excellent corrosion inhibition capability when compared with current chromate-free anticorrosion technology, and comparable performance to chromate-based anticorrosion technology[6,7]. These graphene-based coatings have been used for bipolar plates in fuel cells, which are stainless steel components that separate individual cells in a stack and aid in the distribution of fuel and oxidant, water management and current collection. The high cost of bipolar plates is a limiting factor for the mass-market commercialization of fuel-cell stacks, and ways are sought to reduce their costs or increase their durability, for example by reducing corrosion of the stainless steel in the fuel-cell environment.

Our electrochemical testing indicates that two types of stainless steel with low carbon content, SS 304L and SS 316L, corrode severely in fuel-cell acid environments, at rates of ~0.7 and 0.5 mm yr^{-1}, respectively. When considering the limited thicknesses that can be used for bipolar plates, these corrosion rates are very high. The use of graphene-based coatings resulted in a

significant improvement of the performance of coated stainless steel in the corrosive environment compared with uncoated stainless steel. With a submicrometre graphene-based coating, the corrosion rates of these materials were greatly decreased, to under 0.000196 mm yr^{-1} for SS 304L and under 0.000859 mm yr^{-1} for SS 316L (unpublished results).

But the major concerns of the steel industry over using graphene as a corrosion inhibitor are its high production costs and limited availability. Liquid-phase exfoliation is a cost-effective, environmentally friendly way of producing graphene, in which naturally available graphite is exfoliated by wet-chemical and mechanical processing. The graphene produced by this method, however, has far more defects than chemically synthesized graphene from carbon precursors. Even though the latter suffers from high production cost, it is easier to control the properties of the graphene thus produced, by controlling the process parameters.

In an anticorrosion application, graphene has to be used as one component in a multicomponent coating dispersion. Incorporating graphene into such a coating formulation is still a challenge. Functionalizing graphene can be a solution. The benefit is that the functionalization step can be incorporated during the exfoliation stage, making the whole formulation process a single continuous one.

We foresee that graphene technology will be incorporated into existing processes to enhance products in markets already served by the steel industry. Obviously, new processes will need to be developed for the new graphene-based products, but this is not something the industry will shy away from. ❏

Siva Böhm is at Tata Steel Research & Development (Europe & India), Swinden Technology Centre, Moorgate Road, Rotherham, South Yorkshire S60 3AR, UK, and at IIT-Bombay, Department of Metallurgy & Materials (ME&MS), Powai-Mumbai 400 076, India.
e-mail: siva.bohm@tatasteel.com

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Sequencing with graphene pores

Solid-state nanopores are often used for biomolecular analysis, but have so far been unable to sequence DNA.
Marija Drndić asks whether nanopores made in graphene could fulfil all of the requirements needed for sequencing.

Solid-state nanopores can be used to detect, analyse and manipulate nanoscale objects, and could be used in applications such as nanoparticle analysis and water filtration. They are also of particular interest in the sequencing of DNA. In a typical experiment, a DNA molecule is threaded through a nanoscale hole in a membrane under an applied voltage, and the ion current flowing through this pore is monitored; when the DNA nucleotides enter the pore they block the current by different amounts, and this signal could then be used to identify the individual bases and in turn the DNA sequence.

The field of solid-state nanopores emerged from work on Coulter counters[1], ion channels and protein pores[2–5], and recently proof-of-principle DNA sequencing measurements have been reported using ionic currents and protein pores[6–8]. Compared with protein nanopores, solid-state nanopores offer potential advantages such as robustness, size tunability and compatibility with large-scale chip manufacturing. But solid-state nanopores cannot yet be used to identify individual bases. This is due, in part, to the fact that the membranes used are too thick to probe the local structure of the DNA; they are typically silicon nitride membranes with thicknesses of more than 10 nm, which is equivalent to about 30 DNA bases. As a result, researchers have turned to thin membranes such as graphene, which, because of its atomic thickness, could potentially yield distinct current signals from one or a few DNA bases.

In 2010, three independent research groups — led by Jene Golovchenko, Cees Dekker and myself — reported threading double-stranded DNA molecules through nanopores in suspended graphene membranes[9–11] (Fig. 1). Although there were differences in the experiments — such as whether the graphene was grown[9,11] or exfoliated[12], and the diameters of the nanopores used — the underlying conclusions were similar and showed that the ionic currents could be used to detect the DNA.

In all three of these experiments, the DNA passed through the nanopore too quickly for individual bases to be resolved, and achieving single-base resolution with graphene nanopores remains an unresolved challenge. The percentage of graphene nanopores allowing DNA translocation is also lower than
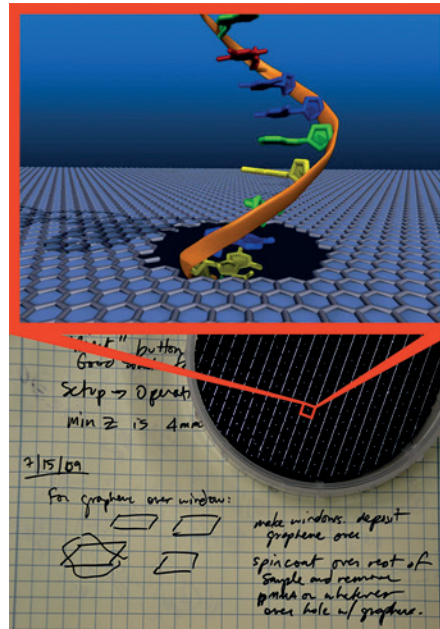

Figure 1 | Top: Illustration of a single-stranded DNA molecule passing through a graphene nanopore. Bottom: Photograph of a solid-state wafer with hundreds of nanopore chips, and laboratory notebook entry[9].

IMAGE COURTESY OF ROBERT JOHNSON

in typical silicon nitride nanopores, mostly owing to wetting problems[9]. Furthermore, more work is needed before graphene nanopores will match the latest achievements of traditional solid-state nanopores made from silicon nitride[12–14] in terms of signal level and signal-to-noise ratio. Nevertheless, the work on graphene nanopores has already inspired a search for other thin membrane materials, and nanopores have, for example, been created from hafnium oxide[15,16], molybdenum disulphide[17] and boron nitride[18] membranes.

For any solid-state nanopores — be it graphene, silicon nitride or something else — there is still a variety of fundamental issues that need to be resolved before DNA sequencing with ionic currents could become a reality. For example, should single- or double-stranded DNA be used? Is a specific range of nanopore diameters required, and how precisely will they have to be manufactured? Improvements in the nanopore devices will also be required. In particular, current noise needs to be lowered

and signal-to-noise ratios increased at high bandwidths[19], or the translocation of the DNA needs to be slowed down without more noise being introduced into the signal. There are also more prosaic but crucial details that need to be dealt with: enough devices have to be tested and reproduced by independent labs to draw firm conclusions, a task that requires skill, time and effort.

At this stage, it is not clear what solid-state nanopore material will be able to meet all of these challenges or, in fact, whether new nanopores will have to be developed to achieve them. There is, however, no theorem stating that only one material will work. More likely, if one works out, others may too — although we should keep in mind that, while it is encouraging that protein pores have shown proof-of-principle sequencing, this is not a proof that any of the solid-state pores will work. And although efforts were undertaken to commercialize nanopores even before any proof-of-principle sequencing demonstration existed, and it is reasonable to extrapolate from existing data, only reproducible experiments can prove a hypothesis. ❑

Marija Drndić is in the Department of Physics and Astronomy, David Rittenhouse Laboratory, University of Pennsylvania, Philadelphia, Pennsylvania 19104, USA.
e-mail: drndic@physics.upenn.edu

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Graphene devices for life

Kostas Kostarelos and **Kostya S. Novoselov** examine the potential of graphene in biomedical applications.
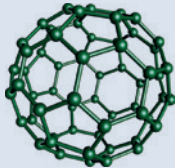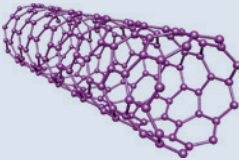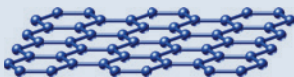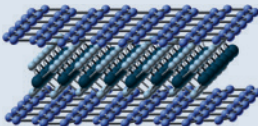
The use of nanomaterials in health and life-science applications has been steadily growing[1,2]. Nanoscience could allow the creation of new approaches in medical intervention, and it promises to revolutionize established clinical practice. The design and engineering of delivery vehicles for diagnostics or therapeutics are just some examples among many; others include point-of-care ultrasensitive sensors, smart (responsive) substrates for artificial tissue design and high-specificity biomarkers. Graphene is considered the archetypal nanostructure within the family of carbon nanoforms, among fullerenes, carbon nanotubes, nanohorns, graphene quantum dots and others. Graphene materials (GMs; a family of materials including pristine graphene sheets, few-layer graphene flakes, graphene oxide

and many others) offer a range of unique, versatile and tunable properties that can be creatively used for biomedical purposes. Their lateral dimensions can be adjusted between nanometres and millimetres, and their thickness can be tuned from single to hundreds of monolayers (also modulating flexural rigidity). The two-dimensional (2D) flat surface can be modified to functionalize the surface character from highly hydrophobic to patchy amphiphilic to highly hydrophilic. The tunability of such properties is perhaps unprecedented among other nanomaterials, offering immense design capabilities. Also important is that graphene can be produced relatively cheaply and in large amounts.

Graphene applications in biomedicine, even though still in their infancy, can be divided into several main areas:

transport (delivery) systems, sensors, tissue engineering and biological agents (for example antimicrobials). The use of various carbon nanoforms in such biomedical applications has been actively pursued for the past decade or more[3]. Therefore, determination of the unique features offered by GMs (and other 2D structures) is of critical importance in the design and development of truly novel constructs of enhanced or previously unattainable functionality. Table 1 illustrates how the most widely explored nanocarbons (fullerenes and carbon nanotubes) compare with graphene and other 2D heterostructures in terms of their biomedical applicability and potential. It also highlights the numerous challenges that each structure faces in implementation and clinical adoption.

Table 1 | Opportunities and challenges in biomedical applications for different forms of nanocarbon.

	Fullerenes	Carbon nanotubes	Graphene materials	2D heterostructures
Unique properties used in biomedical and life science	Free radical scavenging	Cylindrical shape Photothermal capability Inner space (for filling)	2D flat shape Large available surface area Flexibility Electrical conductivity Absence of bandgap Aqueous solubility (in the case of graphene oxide) Versatility of chemical functionalization	Similar to graphene materials in shape and structure More variability in the 'mix' of physical properties from different single layers
Biomedical application (most mature or intensively explored)	Antimicrobials	Molecular transporters (drug delivery) Near-infrared imaging agents	Highly sensitive biosensors Molecular transporters Coatings/substrates for tissue engineering and implants	Almost no such applications reported Biosensing and biodetection will be prime candidates
Opportunity	Interaction with double-stranded nucleic acids (for example mitochondrial DNA)	Translocation of biological membranes and barriers	Responsive to a wide range of parameters High sensitivity Multiple read-out routes Ease and speed of degradation	'Fabricate-by-design' based on the selection of layers
Challenge	Aqueous dispersibility Non-specific DNA binding leading to cytotoxicity	Controlled manufacturing and surface functionalization Aqueous dispersibility Adverse (inflammatory) responses related to fibre shape Slow kinetics of biodegradation	Unknown cytotoxic limitations Controllable dimensions Determination of *in vivo* biodegradability kinetics	Interaction with biological matter is currently unknown

So far, designs of delivery systems based on GMs have primarily attempted to make use of their very large surface-to-volume ratio (larger than with most other materials) and their facile chemical modification. By careful tuning of the dimensions and hydrophobic/hydrophilic surface character of GMs, one can potentially control their capacity to translocate or interact with different biological barriers (for example, membranes or the glomerular filter). Also, the razor-like shape of graphene sheets can potentially help them to slide through membranes and become internalized in a cell. Some evidence of such capacity for particular GM types has been reported[4], but there is still a lack of mechanistic description of the way in which different GMs interact with biological barriers.

It is also possible to consider the graphene flake itself as a therapeutic agent. Research efforts are currently focused on exploring different GM types as local heat sources activated on laser irradiation to ablate tumour tissue by hyperthermia[5]. The absence of a bandgap makes GMs sensitive to irradiation across a wide spectral range, therefore enhancing options for the design of such agents. Antimicrobial activity (bacteriostatic and bacteriolytic) is another biomedical application that is based on the inherent properties of GM sheets themselves, even though there have been contradicting reports about the inherent antimicrobial activity of different GMs[6–8]. For all therapeutic applications in which administration of GMs is envisioned, because the dimensions, surface chemistry and hydrophilicity/hydrophobicity of graphene flakes can vary widely, it will be imperative to understand their impact on some fundamental pharmacological parameters (for example, blood circulation kinetics, immune cell interactions and responses, and tissue distribution) before

they can be tuned to achieve transport to a specific location in the body.

Graphene-based sensor development is another area of application being intensively researched[9]. Because all graphene properties are interlinked (for instance chemical modification will change its electrical resistance, and strain applied to it can be directly detected through the Raman spectrum), it could be possible not only to sense a wide range of parameters, but also to provide a large choice of read-out methods (for instance, chemical doping can be easily detected by transport, optical or Raman measurements). Most promising are the fast biological sensors with either electrical or optical (plasmonic) read-outs in which selectivity is ensured by chemical functionalization. As graphene simultaneously plays the role of a sensing surface (which can be funtionalized to increase selectivity) and the conducting channel, its sensitivity can potentially be very high (note that in many traditional sensors based on field-effect transistors, the conducting channel is separated from the sensing surface)[10]. The other interesting directions in the design of GM-based devices include DNA sequencing through nanopores[11] and the use of graphene as a photodetector or in plasmonic chemical sensors[12]. The maturation and time-to-market of graphene-containing biosensor devices that will not require direct administration into patients are expected to be much more rapid than for administered therapeutics or diagnostics.

Graphene has also been proposed for coating or fabricating matrices and substrates for the engineering of various artificial tissue components and implants. Some of the most promising are artificial skin and orthopaedic implants[13]. Graphene and graphene oxide membranes can also be engineered in different thicknesses and with different chemical functionalities

that could allow controllable permeation of different molecules[14]. It is also conceivable to impregnate graphene-containing membranes with specific (therapeutic) chemical species and release them controllably[15,16].

It should be clear that all these biomedical applications, although actively researched, are still in the early stages of development. A distinction in expectations should be made among different types of device incorporating GMs, mainly determined by the risk–benefit ratio that each particular medical need and application allows[17]. ❑

Kostas Kostarelos and Kostya S. Novoselov are at the National Graphene Institute, University of Manchester, Oxford Road, Manchester M13 9PL, UK. Kostas Kostarelos is also at the Nanomedicine Lab, Faculty of Medical & Human Sciences, University of Manchester, Manchester M13 9PT, UK. Kostya S. Novoselov is also at the School of Physics & Astronomy, University of Manchester, Oxford Road, Manchester M13 9PL, UK.
e-mail: kostas.kostarelos@manchester.ac.uk; kostya@manchester.ac.uk

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Graphene in the sky and beyond

Emilie J. Siochi explains how most of the properties of graphene could be of use in aerospace applications.

With the premium placed on strong, lightweight structures, carbon materials have a long history of use in aerospace applications. Graphitized carbon and carbon–carbon composites[1] are used in thermal protection systems and heat shields, carbon fibre composites are used in aircraft[2], and more recently, carbon nanotubes have been used on spacecraft[3]. As the newest member of this family of materials, graphene also has interesting properties that intersect with unique aerospace requirements. Despite their many attractive aspects, graphene-based structures and systems, like any other material used in aerospace, must clear a number of hurdles before they will be accepted for use in flight structures. Carbon fibre, for example, underwent a development period of several decades between initial discovery and large-scale application in commercial aircraft[4].
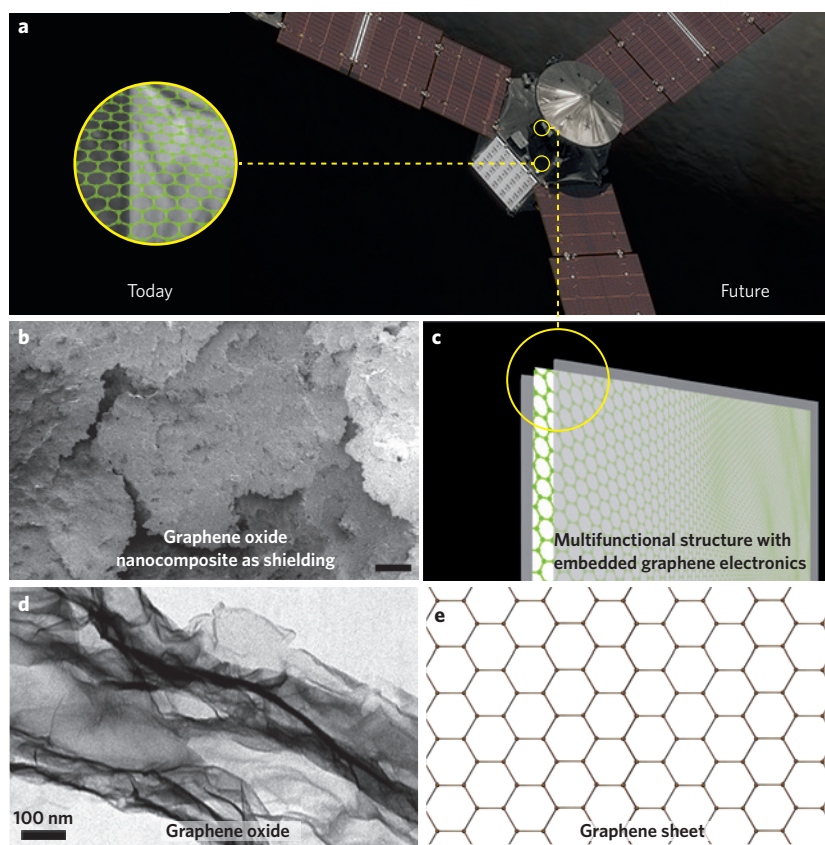
Figure 1 | Notional applications of graphene in spacecraft. **a**, Different parts of a spacecraft could host different types of application. **b**,**d**, A type of application available in the near term. Nanocomposites (**b**) can be made out of graphene oxide (**d**) and used as charge dissipation layers. Scale bar in **b**, 40 µm. **c**,**e**, Future applications in which a graphene sheet (**e**) can be integrated in multifunctional electronic components (**c**). Figures from: **a**, NASA/JPL-Caltech; **b**,**d**, Jae-Woo Kim, National Institute of Aerospace/NASA Langley Research Center; **c**, Susanne Waltz, Lites/NASA Langley Research Center; **e**, © Nicholashan/Thinkstock.

One strategy for accelerating the infusion of new material technologies into aerospace applications is to evaluate the near-, mid- and far-term uses that take advantage of different graphene material properties as they mature during the development process (Fig. 1). For example, although the limited availability of pristine, highly crystalline, single-layer graphene currently constrains its use in electronics to laboratory-scale devices, functionalized graphene is available in commercial quantities. Although some of the exotic electronic transport properties are lost, it retains enough conductivity to function as an electrostatic charge dissipation layer in space structures. To implement this type of near-term application (Fig. 1b,d), the fabrication of nanocomposites containing graphene oxide has to be optimized and then the composites have to be tested to establish if they can replace materials currently used in electrostatic charge dissipation layers. In the mid-term, it would be feasible to envisage the development of high-performance gas-barrier coatings using currently available functionalized graphene. Loss of propellant through boil-off from cryotanks used for in-space propulsion is a significant problem in long-duration missions[5]. Proper dispersion of the two-dimensional graphene nanoparticles in a composite material could greatly reduce propellant loss, as the diffusion path across the barrier material would become so much more tortuous. If the barrier properties exceeded those of state-of-the-art materials based on inorganic clay platelets, the mass reduction could lead to adoption by vehicle designers.

Whereas both the near- and mid-term applications described above are achievable with functionalized graphene nanocomposites whose performance can readily be compared with state-of-the-art materials, the far-term applications are likely to revolve around electronics and will, therefore, require advances in the large-scale production of graphene sheets with very high purity and crystallinity. In particular, the atomic-layer thickness of graphene, along with its mechanical strength and high electron mobility, suggests its suitability for flexible electronics[6]. These devices are amenable to being integrated in multifunctional structures as depicted in Fig. 1c,e. Graphene's potential in this application is broadened when used in combination with MoS_2, another rising star in two-dimensional material systems[7]. The single-layer atomic thickness of graphene has the added benefit of providing intrinsic radiation hardness[8], which can significantly reduce the required mass of conventional radiation shielding material. As in the near- and mid-term application examples, the promise of improved performance and reduced mass makes a case for further research on graphene. In the far-term case, truly revolutionary applications can be envisaged, such as the integration of embedded sensing in multifunctional structures for smart aircraft and spacecraft.

In view of the variety of applications described above, the broader needs of aerospace technology summarized in NASA technology development documents[8] and the possible future uses described in literature reviews[9], graphene has the potential to meet some critical needs in the aerospace arena. Many challenges must, however, be overcome to fully realize its promise. One such barrier is the lack of a large-scale method for reliably and reproducibly producing high-quality graphene. Laboratory-scale demonstrations, although intriguing, are not sufficient to warrant serious consideration in aerospace systems. Maturing a new material requires extensive development, testing, qualification and certification before it can be considered for use on flight vehicles. This is typically done with material that is available in large quantities with minimal batch-to-batch variability in properties. Engineers and designers of air and space-flight vehicles are, sensibly, quite conservative when it comes to adopting new materials, as the environments to which the material will be exposed can be incredibly hostile. They must operate reliably under exposure to both cryogenic and extremely high temperatures, large mechanical and vibrational loads, and a spectrum of radiation. The inherent risk imposed by these environmental conditions and the large investments made in these missions dictate the use of only those materials that meet rigorous qualification and certification standards. Finally, with material selection decisions

typically made early in the design process, the material development process has to align with the time window of the mission development schedule.

The examples of graphene applications for aeronautics and space applications described above are not intended to be comprehensive. Rather, they provide some hints of applications that, in the near term, could enhance current technology solutions, and, in the far term, could enable revolutionary concepts not possible today. Ultimately, the impact of graphene in aerospace applications will depend on reliable functionality, rather than novelty. ❐

Emilie J. Siochi is at the NASA Langley Research Center, Hampton, Virginia 23681, USA.
e-mail: emilie.j.siochi@nasa.gov

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