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Looking at graphene and other 2d crystals in energy conversion and storage

by Francis Sedgemore, 3 February 2015

Scientists working with Europe's Graphene Flagship provide a wide-ranging review of the potential for two-dimensional crystals in energy conversion and storage.

The single-atom thick allotrope of carbon known as graphene has many potential applications, among th energy conversion and storage. Graphene and related two-dimensional crystals combine high electrical conductivity with physical flexibility and a huge surface to weight ratio. Such qualities make them suitable storing electric charge in batteries and supercapacitors, and as catalysts in solar and fuel-cell electrodes.

A number of energy applications for 2d crystals are under development worldwide, and Europe's Graphe Flagship has invested significant resources in this area. In an article for the journal *Science*, flagship scie together with colleagues in the US and Korea, have reviewed the potential for graphene and related mate the energy sector. The authors hope that their review will guide researchers in academia and industry in up a strategy for energy applications and their implementation.

In the *Science* review, the researchers – led by physicist Francesco Bonaccorso, who is based at the Gra Labs of the Instituto Italiano di Tecnologia in Genova, and is a Royal Society Newton Fellow at the Camb Graphene Centre – note the substantial progress made in material preparation at the laboratory level. Th highlight the challenge of producing the materials on an industrial scale in a cost-effective manner.

Physicist Francesco Bonaccorso (photo copyright © 2015 Agnese Abrusci/Instituto Italiano di Tecnologia).

Surface, function and production

Graphene, the best-known of the hundreds of two-dimensional crystals investigated to date, has a very h surface-to-mass ratio. With around 2,600 square metres for every gramme, graphene is all surface and n and it is this 2d nature which gives graphene its unique electrical, thermal and mechanical properties.

Other two-dimensional materials, including the transition metal dichalcogenide molybdenum disulphide (M various transition metal oxides, and the MAX-phase class of 2d crystals, display properties complementa those of graphene. In all cases, the potential for mass production and chemical functionalisation make gr and related materials an ideal platform for energy applications. The challenge is to produce them on a lar with properties tailored for specific purposes.

There are various ways of producing graphene in bulk, but, when it comes to industrial scale manufacture of the volumes required for energy applications, liquid-phase exfoliation looks very promising. This involves exploiting the agitating effect of high-frequency sound waves to separate graphene flakes from graphite held in suspension. Electronic-grade graphene can be produced through chemical vapour deposition, but the process is less suitable for energy applications. A third possibility is the chemical synthesis of graphene flakes, but there are questions of scalability.

Solar cells

Barely a week goes by without reports of new developments in solar cell technology. Efficiencies are on the rise, and various nanomaterials are employed in the manufacture of photovoltaic films and electrodes. Silicon remains the most widely used photon absorber, and this established semiconductor material dominates the solar cell market. Second and third-generation photovoltaic cells, including those based on organic materials and quantum dots, have lower photocurrent efficiency than those fabricated from silicon, and there are issues with material stability and durability.

Graphene and related materials are attracting serious interest in solar energy conversion, on the grounds that they could improve efficiency, reduce production costs, and deal with a number of environmental issues. In addition, 2d crystals have the advantages of transparency and flexibility, and in time they could replace indium tin oxide films, which are relatively brittle. They can also act as catalysts in dye-sensitised solar cells, in place of platinum. This may result in a four orders-of-magnitude reduction in device cost.

Graphene can be engineered to match the low sheet resistance and high transmittance of transparent conducting films made from established materials, and graphene-based materials have with varying degrees of success been implemented in solar energy systems, including inorganic, organic, dye-sensitised and hybrid organic/inorganic cells. This is an especially active area of research and development, with chemically-doped graphene a particular focus of attention.

As well as the photo-sensitivity of solar cells, charge collection and transport are important issues to consider, and this applies both to dye-sensitised cells and organic photovoltaics. For example, graphene and related materials can be used to lessen the negative effects of charge carrier recombination. In the case of dye-sensitised cells, reduced graphene oxide is employed in combination with titanium dioxide nanoparticles, whilst graphene quantum dots are efficient electron hole transport layers in organic photovoltaics.

There is a need to replace or at least reduce the amount of platinum in solar cell counter-electrodes, owing to the cost of the precious metal, and its tendency to degrade when in contact with iodide electrolytes, which reduces device efficiency. Functionalised graphene flakes and other 2d crystals have been used in place of platinum in counter-electrodes, sometimes outperforming the platinum.

The efficiency of photovoltaic devices based on graphene and related materials is improving at a pace superior to those based on established materials, with the highest reported efficiency of 13% for dye-sensitised cells achieved using graphene nanoplatelets as a counter-electrode. With graphene-based perovskite solar cells we have seen an efficiency of 15.6%, and that achieved with relatively low-temperature processing, yielding a significant cost reduction.

Thermoelectric devices

Recovering waste heat is clearly a good thing, and this may be done with solid-state devices which generate electricity from temperature gradients. Thermoelectric devices can also convert heat produced by sunlight into electricity, and here they can enhance solar cells by capturing heat produced by photons with energies below the band gaps of the photosensitive materials employed.

The effectiveness of thermoelectric devices is assessed by the fraction of absorbed heat converted into electricity, along with a figure of merit which depends on the material's electrical and thermal conductivities. Traditional thermoelectric devices have conversion efficiencies of around 5%, and this low value limits their widespread use.

In thermoelectric materials one is looking for high electrical and low thermal conductivity. On the face of it this rules out graphene, in which the electrical and thermal conductivities are both high. However, it is possible to tailor the thermal transport properties of graphene by introducing defects, edge roughness, and periodic holes into the material. This can reduce thermal conductivity by up to 100 times when compared with pristine graphene. It can also raise the figure of merit by a factor of three over that obtained with other materials.

The problem here is the difficulty in scaling tailored graphene nanoribbons via chemical synthesis. Still, we can blend two-dimensional materials with carbon nanotubes to increase electrical conductivity without reducing thermoelectric sensitivity.

Fuel cells

A fuel cell converts chemical energy from a fuel such as hydrogen gas into electricity via a reaction with oxygen or another oxidising agent. Much of the attention devoted to fuel cells focuses on high power applications.

When it comes to improving and better exploiting fuel cells, the principal difficulty is one of cost, and specifically the need to replace expensive noble metals such as platinum and gold as catalysts in the chemical reaction. Fuel cell electrodes must also be chemically stable over the long term, and for some applications physically flexible.

Graphene and related materials are promising candidates for fuel cell electrodes, and also as membranes in proton exchange fuel cells. Where they cannot replace precious metal catalysts, the addition of two-dimensional materials may lower the amount of expensive catalyst materials required. For example, reduced graphene oxide modifies the properties of platinum electro-catalysts supported on it, leading to improved methanol oxidation when compared with commercial platinum/carbon black mixtures.

Batteries

Today's state-of-the-art rechargeable batteries are based on lithium-based cathodes and graphite anodes, and crucial to their performance is the capacity of the anode material to store lithium ions. Graphene has a larger gravimetric capacity than graphite, and the flexible nature of the material has advantages when it comes to certain applications and environments. Graphene and related materials are also appealing as fuel cell cathodes, owing to their high electrical capacity per unit weight.

Graphene-based hybrid electrodes may also be employed in rechargeable batteries to increase electron transport, capacity, discharge current and device longevity. Where graphene is not actually incorporated into battery electrodes, it may be used as a substrate for the growth of high-performance anode/cathode nanoparticles. For example, lithium-based nanorods grown on reduced graphene oxide flakes show a significantly lower degradation over a fixed number of cycles than reduced graphene oxide or graphite. Wrapping electrochemically-active particles within graphene or MoS2 flakes is another possibility.

Supercapacitors

Electrochemical capacitors store energy in an electric field set up between conducting plates separated by an insulating material. Supercapacitors, which divide into electrostatic double-layer capacitors, hybrid and pseudocapacitors, are ideal for high power applications in which the required energy density is an order of magnitude greater than is possible with lithium-ion batteries. Such applications include electric and hybrid motor vehicles, heavy lifting, load levelling and backup power for electric utilities and industrial plant.

Supercapacitors are necessarily large devices, and the materials of which they are made are produced in bulk into electrodes 100 to 200 microns thick. Most double-layer capacitors have carbon electrodes impregnated with organic electrolytes. Another type of supercapacitor is based on lithium-ion hybrid cells, in which a graphite lithium-ion anode is coupled with an activated carbon cathode. In this case the energy density is around twice that of double-layer capacitors.

The performance of supercapacitors is dependent on the electrode surface area accessible to the electrolyte, and this is where 2d crystals such as graphene have the advantage over graphite and activated carbon. In practice this can include graphene-based platelets with spacer materials such as carbon nanotubes, mesoporous carbon spheres, water and ionic liquids, and resins chemically activated to create a porous structure.

Note that large material surface areas do not always translate into high-performance supercapacitors, especially if the material packing density is low. It is possible to increase the packing density through evaporation drying of graphene hydrogels, and by capillary compression of reduced graphene oxide. Raising the operating voltage is another way of increasing the energy storage capacity.

Double-layer capacitors are marked by their rapid charge/discharge and long cycle life, and lithium-ion batteries by a high energy storage capacity. Combine the two qualities, and the result is a lithium-ion hybrid supercapacitor, albeit with performance trade-offs common to both types of device.

Electrodes for double-layer capacitors made from microwave-expanded graphite oxide and lithium-ion battery electrodes comprising graphite, lithium and iron oxides have been studied, as have electrodes containing metal oxides based on ruthenium, manganese and molybdenum, with conducting polymers to increase the specific capacitance via chemical reduction and oxidation reactions. Graphene is included in these systems as a conductive support.

Hydrogen production and storage

Hydrogen has an energy density more than three times that of petrol, and its combustion by-product is water. The principal challenges in using hydrogen as a green fuel are to produce and store the gas.

Electrolysis is the key mechanism for hydrogen gas production, and the edges of 2d crystals including MoS2 and reduced graphene oxide are active catalytic sites in the reaction. Whilst simple in outline, the detail of the hydrogen evolution reaction varies from material to material, with resistive losses a significant issue with non-metallic electrodes. Combining graphene and related materials with carbon nanotubes can enhance the reaction by improving electron transport efficiency.

When it comes to storing hydrogen, carbon-based structures are attractive owing to their high gravimetric capacity, and graphene particularly so. Calculations show that the gravimetric density may in principal be up to 8% in graphene multi-layers spaced with pillar structures such as carbon nanotubes. This applies to cryogenic temperature and/or high pressure regimes. With ambient conditions the maximum theoretical density is 4%. In practice the numbers are lower, with up to 1% density at room temperature and pressure.

Doping graphene with alkaline or transition metals can increase the gravimetric density to 10%. This can be achieved by functionalising graphene with metals such as palladium, which catalyse the dissociation of hydrogen molecules into ions on the graphene surface.

Perspectives

Graphene and related two-dimensional crystals may play a major role in future energy conversion and storage technologies, and this is an active area of research and development for Graphene Flagship partners, both academic and industrial.

"The huge interest in two-dimensional crystals for energy applications comes both from their physico-chemical properties, and the possibility of producing and processing them in large quantities, in a cost-effective manner," says Bonaccorso. *"In this context, the development of functional inks based on two-dimensional crystals is the gateway for the realisation of new generation electrodes in energy storage and conversion devices."* Bonaccorso adds that the challenge ahead is to demonstrate a disruptive technology in which two-dimensional materials not only replace traditional electrodes, but more importantly enable whole new device concepts.

Review co-author Andrea Ferrari, who chairs the Executive Board of the Graphene Flagship, and is director of the Cambridge Graphene Centre, offers a soberly optimistic view of the potential for graphene: *"Graphene and related materials have great promise in these areas, and the Graphene Flagship has identified energy applications as a key area of investment. We hope that our critical overview will guide researchers in academia and industry in identifying optimal pathways toward applications and implementation, with an eventual benefit for society as a whole."*

Francis Sedgemore is the science writer for the Graphene Flagship.

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