# Graphene Battery User's Guide

This Graphene Battery User's Guide, which has been created for scientists and non-scientists alike, details how graphene batteries work, their benefits, and provides immediate, actionable steps that you can take to begin developing your own graphene battery. Don't miss out on the next phase of nano evolution. Our Graphene Battery User's Guide includes four well established graphene electrode designs of experiments (DOE) from cutting edge academic research including one for a Graphene-Lithium-Sulphur battery, the current front runner technology, and are included for reference.
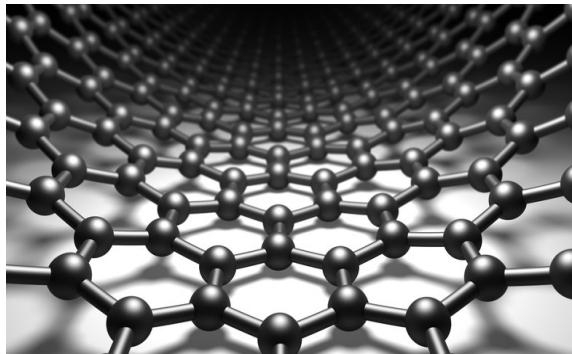
Above – A graphical representation of a graphene sheet

Graphene-based batteries are being actively researched for many commercial applications. The improved performance and life cycle advantages when developing graphene-based batteries over traditional metal-ion batteries are well worth the resource investment. Elon Musk's Tesla Motors provides a famous example of innovative companies actively pursuing graphene battery research and commercialization. We believe that the real graphene battery breakthroughs will be from graphene-lithium-ion hybrid chemistries incorporated into the cathodes of lithium-sulfur cells. This type of technology is still years away from commercialization and intensive research is ongoing. The more innovative graphene battery technologies will require significant R&D expenditures and will take many years to commercialize.

To learn more about the types of Graphene, synthesis methods, properties, and applications please see our Graphene User Guide.

Traditional Battery Technology

Above- An cross sectional view of a traditional battery

Batteries, whether they are rechargeable are not, commonly employ lithium. Zinc and alkaline based batteries are available, but they generally have a shorter lifespan due to their high charge density. Unlike lithium-based batteries, they can't operate at higher voltages. A primary (non-rechargeable) battery consists of two electrodes which allows the current to flow in one direction only, via an intermediary electrolyte. Secondary (rechargeable) batteries still consist of two electrodes however the lithium ions can flow in both directions depending on if charging or discharging. The anode is generally a lithium-based (metal oxide) compound and the cathode a porous carbon. Both the cathode and anode have a rigid structure with defined holes, which allows for the absorption of lithium ions into the holes when the current is applied. When there is no current being applied, the ions desorb into the electrolyte solution. Absorption of the lithium ions can occur on both the anode and the cathode. When a battery is in use, the ions move to the cathode. When charging, the current is reversed and the ion absorb into the anode. This process allows for many cycles to be produced, leading to an enhanced lifespan. The cathode of choice is traditionally graphite and the anode can vary, but common types are $LiCoO_2$, $LiMn_2O_4$, $LiNiMnCoO_2$ (NMC), $LiFePO_4$, $LiNiCoAlO_2$ and $Li_4Ti_5O_{12}$.

Graphene Battery Technology

Graphene Batteries can reduce the environmental impact of battery use

Graphene battery technology has a similar structure to traditional batteries in that they have two electrodes and an electrolyte solution to facilitate ion transfer. The main difference between solid-state batteries and graphene-based batteries is in the composition of one or both electrodes. The change primarily lies in the cathode, but carbon allotropes can be utilized in the anode as well. The cathode in a traditional battery is purely composed of solid-state materials, where as in a graphene battery the cathode is a composite-a hybrid material consisting of a solid-state metallic material and graphene. The amount of graphene in the composite can vary, depending upon the intended application. The amount of graphene incorporated into the electrode generally depends upon the performance requirements and is based upon the existing efficiencies and/or weaknesses of the solid-state precursor material.

Graphene Battery Breakthrough

The real graphene battery breakthrough are the graphene-lithium-ion hybrid chemistries incorporated into the cathodes of lithium-sulfur cells as detailed in this guide. There are no pure graphene electrodes in a graphene battery, many graphene-based electrodes are fabricated and work in a similar way to traditional batteries. Their performance is enhanced via the addition of graphene to the electrode formulation. Generally, inorganic-based electrodes will have limitations which are typically surface area, density, capacity, cycle times, conductivity or capacitance to name a few. As graphene is a

versatile molecule with many unique and desirable properties, it can be adopted in a variety of ways as there is no 'one size fits all' solution for using graphene. Graphene is used to enhance many of the benefits already present with traditional materials, but it also helps to breakthrough previous battery limitations, leading to increased battery performance or life. Graphene works in electrodes in two general ways, either as a support or a composite/hybrid. As a support material, graphene helps to keep metal ions in a regular order, which generally helps with electrode efficiency. As a composite material in an electrode, it plays a different role as they are generally more involved in the facilitation of the charge itself, where its high conductivity and well-ordered structure are critical to providing an improvement against it's non-graphene predecessors.

Below is a detailed account of how graphene specifically enables certain battery applications.

Lithium-Ion Batteries

Graphene-based batteries are quickly becoming more favorable than their graphite predecessors. Graphene batteries are an emerging technology which allows for increased electrode density, faster cycle times, as well as possessing the ability to hold the charge longer thus improving the battery's lifespan. Graphite batteries are well-established and come in many forms. Similar to graphite, there are now various types of functional graphene derivative electrodes and researchers are discovering multiple benefits when compared to pure graphite electrodes.

Graphene-Metal Oxide Hybrids

As mentioned, graphite has been historically used as the primary cathode material, where the lithium ions migrate into the structured holes. However, graphene doesn't have this capability. Instead, due to the large surface area of graphene, the lithium ions can be stored via surface adsorption and induced bonding. Induced bonding generally occurs when a graphene derivative is present and the lithium ions bind to the functionalized surface. In addition to a large surface area, graphene electrodes also possess high conductivity. Many of the metal oxides traditionally used in batteries have impairments such as low conductivity, low volumetric energy density and the loss of contact points. By hybridizing the metal oxide matrix with graphene, many of these problems are removed and the conductivity becomes greater because the interaction between the interstitial ions and the hybrid matrix is vastly improved. To produce Graphene-Metal Oxide nanoparticle hybrids, the graphene acts as a template during the synthesis which produces an evenly distributed matrix due to the regular repeating structure of graphene. This process also limits nanoparticle aggregation which promotes the large nanoparticle surface area during the lithium charge and discharge cycles. As such, the specific capacity and cycling

performance is improved against pure MO-based electrodes. Graphene-MO hybrid electrodes can exhibit up to 1100 mAh g^{-1} for the first 10 cycles. The specific energy density is maintained at 1000 mAh g^{-1}, even after 130 cycles.

Graphene-Carbon Nanotube/Fullerene Hybrids

While not as widely reported, an electrode consisting of both graphene and either carbon nanotubes and/or fullerenes have been implemented. Electrodes that consist of 6-15 monolayers of graphene, which are layered on top of each other, show a specific capacity of 540 mAh g^{-1}, which is a vast improvement on their graphitic counterparts due to their higher surface area. By dispersing the graphene sheets with either carbon nanotubes or fullerenes, the inter-graphene spacing is increased. This extra spacing creates extra cavities for the lithium ions to occupy and can increase the specific capacity by up to 40% when compared with stacked graphene electrodes.

Graphene Lithium Sulphur Batteries
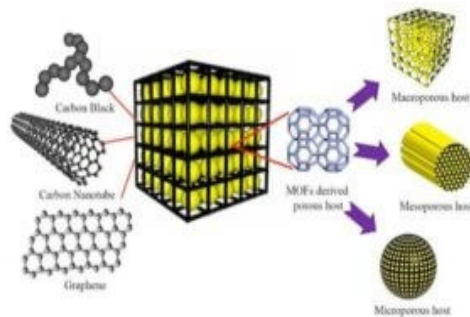

Above- a Graphene-Lithium-Sulphur Battery

Lithium sulphur batteries have the potential to replace lithium-ion batteries in commercial applications due to their low cost, low toxicity and the potential for possessing an energy density of 2567 W h kg^{-1}, which is five times than that of lithium-based batteries currently available. As such, they have attracted a lot of interest. However, lithium sulphur batteries have several significant drawbacks that have stopped them from reaching the commercial marketplace. The first is the deposition of inorganic salts at the cathode, due to the cell possessing highly soluble reactants. The deposition of the salts causes a

loss in active material, low coulombic efficiency, low utilization of the sulphur cathode and a degradation of the cycle life. The second main concern is that sulphur suffers from an inherent low conductivity.

Using graphene as a support for the sulphur ions eradicates some of the associated problems in sulphur batteries due to many properties previously mentioned; it's high electrical conductivity, good mechanical strength, high chemical/thermal stability and a large surface area. The high surface area provides a good dispersion of sulphur, which regulates the mobilization of sulphur ions and prevents a build-up of sulphur ions on the cathode. The production of graphene supported sulphur particles can also be produced through a one-pot synthesis.

The performance of lithium-sulphur batteries depends heavily upon the concentration of sulphur (and effectively the graphene to sulphur ratio) in the electrode. From electrodes produced to date, the higher sulphur containing electrodes provide the best performance. Electrodes containing 63 wt% sulphur produce a capacity of 731 mAh g^{-1}, with the potential of obtaining up to 1160 mAh g^{-1}. After 50 cycles, the capacity is maintained at 700 mA h g^{-1}. This is at least twice the capacity of other sulphur-graphene electrodes produced and they are also stable under high rate cycling. Some degradation of the cell does occur as the sulphur binding has not yet reached 100% efficiency. Nonetheless, graphene supported sulphur electrodes show a vast improvement when compared to non-graphene-based sulphur electrodes.

Graphene Supercapacitors
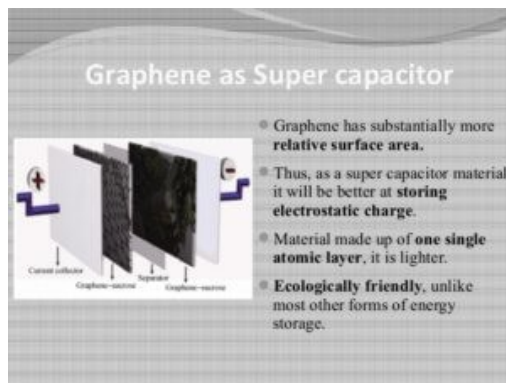
A Graphene Supercapacitor Design

Supercapacitors are a valuable commodity in the field of electronics and can store up more than a hundred times more energy than standard capacitors. Supercapacitors are also able to work in low temperature conditions and are regularly used in lieu of electrochemical batteries.

One of the main properties of a supercapacitance material is the ability to form double-electric layers. This is essential in electric double-layer capacitors *(*EDLC) supercapacitors. Supercapacitors work by accumulating charges at the electrode-electrolyte interface through polarization, so that the energy is stored. Activated carbon has been the traditional electrode choice, but it suffers from the inability to operate at high voltages. Graphene, and its derivatives, are useful due to their high specific surface area, high conductivity, open-pore structure, production potential and low cost; all of which are desirable properties for a supercapacitor.

# Graphene-Metal Oxide Composite Electrodes
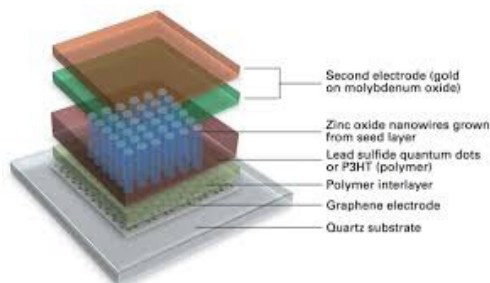
Graphene Molybdenum Oxide Electrodes

Graphene and metal oxide composite electrodes have provided a new area in which the capacitance is increased compared to standard capacitor electrodes. Common metal oxides include ZnO_2, SnO_2, Co_3O_4 and MnO_2. MnO_2 graphene composites are the most promising due to the variable oxidation states that manganese ions can adopt. The redox reaction between the III and IV states involves the intercalation of metal ions in the electrolyte solution (e.g. Li^+, Na^+), which helps to facilitate energy storage. In addition to the efficient ion intercalation, the graphene sheets yield a conductive network with a large surface area, which helps to further promote ion-electrode interactions. Graphene-MO composites show a high specific capacitance of 310 F g^{-1}, which is three times higher than an electrode made of either pure graphene or metal oxide. 95.6% of the specific capacitance can also be retained after 2000 cycles when using graphene-MO composites.

As mentioned, supercapacitors are currently being favored over electrochemical batteries for some applications. However, one issue with using supercapacitors is the lower energy density. Graphene has been incorporated into the negative electrodes of supercapacitors to increase the energy density, while keeping the power density high. Supercapacitors have been developed using graphene as the negative electrode and a nanowire-graphene composite as the positive electrode. This capacitor is set up asymmetrically and has found to give a high-energy density of 30.4 Wh kg^{-1} at an operating voltage of 2 V. The graphene supercapacitors have surpassed other capacitors in terms of both energy and power density and is a promising prospect in the field of supercapacitors.

Graphene-Polymer Composite Electrodes
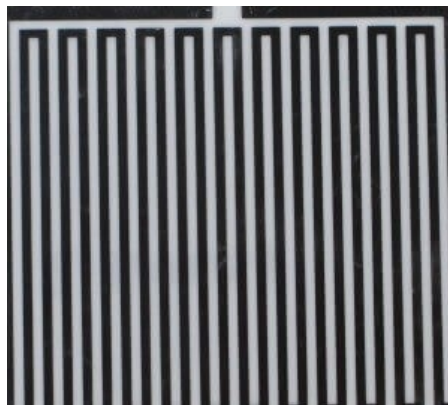
A Graphene-Polymer Composite Electrode

While graphene-polymer composites don't possess high conductivity when compared to other graphene-based composites, they do possess a high charge/discharge rate, flexibility, and high doping-undoping capability. Graphene-polymer composites operate by n and p doping redox reactions where electrons are lost/gained to convert and store energy. Graphene oxide and a nitrogen containing polymer are best used to create a graphene-polymer electrode composite. Polymerization of the functional groups facilitates strong pi-pi interactions between the two components of the composite which leads to a large surface area and a semi-flexible structure that can mechanically deform during the cycle charge-discharge processes. These graphene-polymer composites can exhibit up to 531 F g^{-1} and retain up to 74% of its capacitance after 2000 cycles.
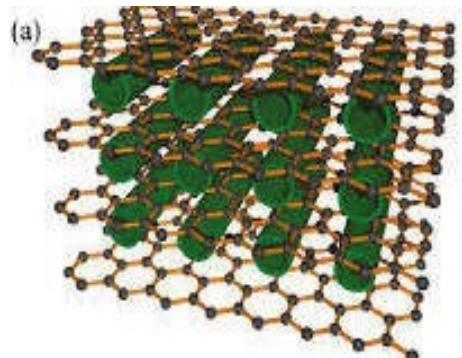
Graphene-CNT Electrodes

A representation of aligned carbon nanotubes in between graphene sheets

Like lithium-ion batteries, carbon nanotubes are used to increase the inter-graphene spacing between the various monolayers. Stacked graphene electrodes can suffer from a decreased surface area compared to monolayered graphene. By introducing nanotubes into the electrode, both the nanopores and the conductivity of the electrode are increased. Nanotubes can be employed into the graphene matrix in either two or three-dimensions, with an average specific capacitance of 120 F g^{-1} and 386 F g^{-1}, respectively. In the three-dimensional graphene-CNT, the capacitance can even increase by up to 20% after 2000 cycles, showing that these electrodes have an excellent electrochemical stability.

Graphene Fuel Cells
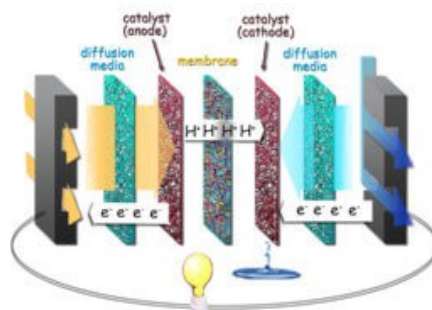
A Fuel Cell Inside View

Fuel cells are different from batteries and capacitors, as they produce electricity rather than store it. Many fuel cells contain a platinum-based catalyst, which are very expensive to produce. To minimize the cost of fuel cells, carbon allotropes are used as supports for the platinum catalysts. Graphene is one such catalyst support. Graphene oxide provides a good dispersion, large surface area and high conductivity (when reduced), so the integrity and efficiency of the device is enhanced by integrating graphene into the composite materials used in fuel cells.

Graphene is used in some fuel cells to help facilitate the oxidation of methanol and has been found to be much more effective than other carbon allotropes, such as carbon nanotubes and carbon black. Graphene's two-dimensional sheets provide a greater active surface area for electron/ion transport, as both sides of the sheet are exposed to the solution within the fuel cell. The uniformity of the graphene surface also prevents aggregation and promotes an even distribution of the platinum particles across the supports. The surface defects in the graphene structure also increases the interaction between the graphene support and the platinum particles. Platinum-graphene supported fuel cells can exhibit a current density up to 0.12 mA cm^{-2}, which is at least three times higher than other carbon-based supports.

Graphene can be doped with nitrogen after undergoing a nitrogen plasma treatment. Doped graphene has nitrogen-based functional groups on its surface which allow for a better dispersion and decoration by the platinum nanoparticles. Doped graphene produces a greater conductivity and electrocatalytic activity compared to undoped graphene. The oxidation current of doped graphene is twice that of undoped graphene.

Design Of Experiments- Synthesizing A Graphene-Based Battery Electrode

The conversion of graphene and metal-based inorganic compounds into usable graphene composites for electrodes can be done by many methods. As this new field is advancing, new methods are frequently being invented and subsequently published. To date they include *ex-situ* hybridization, *in-situ* crystallization, chemical reduction, electroless deposition, sol-gel methods, hydrothermal methods, electrochemical deposition, thermal evaporation and *in-situ* self-assembly, to name a few of the most common classes utilized.

As with any method, there are always multiple ways to synthesize the material itself. To cover each one would be impractical, so here we look at some specific ways that you can implement graphene into composite materials for use as electrodes in your graphene battery R&D project. The values used are from published experiments and are used for ratio illustrative purposes only. The amounts and scale can be varied to better suit specific experiments.

Below is a DOE for Graphene-Lithium-Sulphur batteries, a current leading technology.

"Synthesis of the Thermally Exfoliated Reduced Graphene. Thermally exfoliated reduced graphene was obtained.

Preparation of Graphene-Sulphur Hybrids- The G/S hybrids were prepared by hydrothermal reduction assembly of GO with a sulfur-dissolving CS_2 and alcohol solution. In brief, 50 mL of the GO aqueous dispersion and 15 mL of alcohol were mixed, and then 3 mL of CS_2 containing 100, 150, and 200 mg of dissolved sulfur (tuning the sulfur content in the samples) was added to the GO dispersion. The mixture was stirred for 90 min and then sealed in an 80 mL Teflon-lined stainless steel autoclave for a hydrothermal reaction at 180 C for 10 h. The black cylinder of the G/S hydrogel was washed by ethanol and distilled water, and the wet hydrogel was then freeze-dried to obtain the G/S hybrids.

Preparation of Graphene-Sulphur Hybrids (Powder)- G/S hybrids (powder) were prepared by mixing 90 mg of intercalation-exfoliated graphene and thermally exfoliated reduced graphene with 150 mg of sulfur under the same hydrothermal conditions as the G/S hybrids.

Preparation of Graphene-Sulphur$_{mix}$ The G/S$_{mix}$ was prepared by mixing- 50 mL of the GO aqueous dispersion, 15 mL of alcohol, and 150 mg of sulfur under the same conditions but without CS_2.

Electrochemical Measurements. The G/S hybrid was cut, compressed, and shaped into a circular pellet with a diameter of 12 mm and directly used as a cathode. The mass loading of a G/S electrode was about 2 mg cm^2. The G S59 or G S60 hybrid (powder) cathode was prepared by mixing 90 wt % G S59 or G S60 hybrid (powder) with 10 wt % polyvinylidene fluoride dissolved in N-methyl-2-pyrrolidone as a binder to form a slurry, which was then coated on an aluminum foil and dried under vacuum at 70 C for 12 h. The foil was pressed between twin rollers, shaped into a circular pellet with a diameter of 12 mm, and used as a cathode. The electrolyte was 1.0 M lithium bis-trifluoromethanesulfonylimide in 1,3-dioxolane and 1,2-di- methoxyethane (1:1 by volume) with 0.5 wt % LiNO$_3$ additive. A 2025 type stainless steel coin cell was used to assemble a test cell. A lithium metal foil was used as the anode, and a G/S slice as the cathode. A LAND galvanostatic charge discharge instrument was used to perform the measurements. The coin-type cell was assembled in an Ar-filled glovebox (MBraun Unilab). The current density set for cell tests was referred to the mass of sulfur in the cathode and varied from 0.3 to 4.5 A g^{-1}. The charge discharge voltage range was 1.5 2.8 V. The CV was measured using a VSP-300 multichannel potentiostat/galvanostat (Bio-Logic, France) workstation in the voltage range 1.5 2.8 V (vs Liþ/Li) at a scan rate of 0.1 mV s^{-1}. The G S63 hybrid electrode was discharged to the end of the second plateau and disassembled, dried in the glovebox, and followed by transferring to the vacuum

chamber of XPS for structure characterization."[1]

Below are 3 more types of Graphene Electrode DOE's

To create a pure graphene-based electrode, disperse graphene oxide powder (100 mg) in distilled water (30 mL) and sonicate for 30 minutes. Heat the resulting suspension on a hot plate until it reaches 100 °C and add hydrazine hydrate (3 mL). Keep the suspension at 98 °C for 24 h to reduce the graphene oxide to rGO. The reduced graphene oxide can be collected by filtration to leave a black powder. Wash the filtrated powder several times with distilled water, so that the excess hydrazine is removed. Re-disperse the graphene powder into water by sonication. Centrifuge the solution (4000 rpm, 3 minutes) to remove the larger particles. Collect the graphene by vacuum filtration and dry in a vacuum. If you have purchased rGO, then this step can be skipped. To create the electrode, disperse graphene in ethanol until a concentration of 0.2 mgmL^{-1} is achieved. Filter the suspension by vacuum filtration and collect on the microporous filter paper. Cut the filtered graphene into 1 x 2 cm^2 (1 mg weight), so that it is ready for use. Attach it to a cell, with an electrolyte buffer, to test the graphene electrode.

This second method details the preparation of a cobalt-graphene hybrid electrode, for use as an electrode in lithium-ion batteries. To prepare the electrode, add graphene oxide (0.1 g) to cobalt acetate (350 mg) and deionised water (400 mL). To the solution add NH_4OH (3800 µL) and hydrazine (250 µL) and stir for 4 h at 100 °C. Filter the solution once the reaction has finished. Re-crystallize the solution by heating the product for 6 h at 200 °C.

This final method is for creating a tin-graphene nanoribbon composite electrode, for use in lithium-ion batteries. To create the electrode, add graphene nanoribbon (GNR) (75 mg), $SnCl_2.H_2O$ (1.33g, 5.89 mmol), 2-pyrrolidinone (65 mL) and a magnetic stirrer bar to a dried round-bottom flask. Sonicate the solution for 20 minutes, followed by refluxing for 1 h. Cool the vessel to room temperature and sonicate overnight in an open-air environment. Quench the mixture with acetone and water three times and filter over a PTFE membrane (0.45 µm). Dry in a vacuum (60 °C) for 24 h and anneal in a quartz furnace (500 °C, Ar atmosphere) for 2h. The theoretical yield is 380 mg.

Graphene Products Available

Cheap Tubes Inc offers various graphene-based products that can be implemented into electrode formulations.
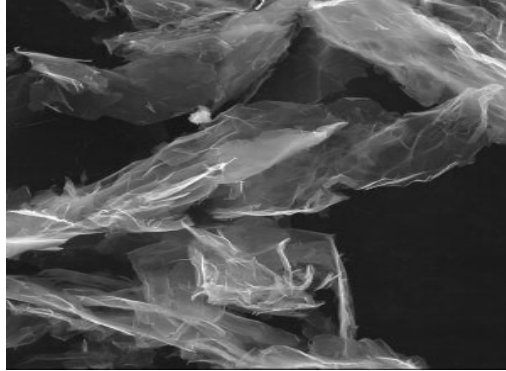
Above- Our Plasma Exfoliated Graphene Nanoplatelets

Graphene Nanoplatelets (GNPs) offer some of the best properties for battery applications. The GNPs can be used to replace other carbon-based materials. GNPs have excellent electrical and thermal conductivity, mechanical stability and can provide a composite with enhanced conductivities, mechanical strength, and lower gas permeation. Our research grade graphene nanoplatelets are produced by plasma exfoliation. Our plasma exfoliation process produces high-grade GNPs which have less defects and a higher internal conductivity. The GNPs consist of several graphene layers and generally have a thickness between 3-10 nm and are friable with high shear methods such as a 3 roll mill or homogenizer. The GNPs are available with varying functional groups, including nitrogen-based, oxygen-based, fluorine and amine groups. Non-functionalised GNPs (argon processed) are also available. The production methods are scalable, so GNPs can be implemented in large volume and larger scale applications.

Graphene Oxide (and reduced graphene oxide) is available as a powder, a dispersion, or a spin coated film. The elemental composition of Cheaptubes graphene oxide consists of 35-42% carbon, 45-55% oxygen and 3-5% hydrogen. Dispersions can be provided in various solvents and in a range of concentrations. We also offer graphene oxide (and reduced graphene oxide) films and coatings. A graphene oxide film comprised of our single layer graphene oxide product spin coated on glass provides a 5-20 nm final thickness. They also possess a conductivity in the range of 10^4-10^5 Sm^{-1}, with a sheet resistance of 10^1-10^3 Ωsq^{-1}. In contrast, a single (flexible) graphene sheet on a flexible organic substrate provides the same thickness and area, but it provides a conductivity in the range of 10^3-10^4 Sm^{-1} and a sheet resistance of 10^2-10^4 Ω sq^{-1}.

CVD Graphene Films are also available. Graphene films that have been grown on a range of substrates

including silicon, copper, PET and quartz, are available. They can also be transferred to a variety of customer supplied substrates if they are compatible with the solvents used to float the graphene off the substrate so it can be redeposited onto your substrate.

As an alternative to graphene, conductive carbon nanotube composite additives can be used. This product has been specifically designed to improve the efficiency of lithium-ion electrodes. As a blend of both nanotubes and carbon black, it improves the tap density (up to 10%) and capacitance retention of the electrode without decline after multiple charge/discharge cycles. The additive contain multi-walled carbon nanotubes blended with a proprietary carbon black. This additive can be used in both the anode and cathode within a battery cell. Generally 2-3 wt% is used in a cathode and 1-2 wt% in an anode.

Conclusion

Graphene-based batteries are quickly becoming comparable, in terms of efficiency, to traditional solid-state batteries. They are advancing all the time and it won't be long before they surpass their solid-state predecessors. The extra benefits associated with graphene being present in the electrodes can be useful, even if the efficiency isn't as high. For batteries that possess a similar efficiency, graphene batteries are an ideal choice, which is why scientist are trying to further advance this class of batteries. They have started to gain traction in the commercial marketplace and it won't be long before they become the norm and phase-out solid-state batteries. To quote recent forecasts "the world graphene battery market is expected to reach $115 million by 2022, growing at a CAGR of 38.4% during the forecast period. The automotive industry is estimated to dominate the market throughout the analysis period. Geographically, Europe is expected to be the leading market in 2016, with a revenue contribution of around 38%."

With increasing energy demands globally, improving energy storage devices while reducing negative environmental impacts related to consumer based battery usage is a noble goal and one that we emphatically support. We hope that this guide has helped you to understand the current graphene battery research trends and inspired you to begin graphene battery development.

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