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Marriage of Top-Down Lithography to Bottom-Up Chemistry Edge Control in Graphene Nanoribbons

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14. ABSTRACT

In this reporting period we have continued to explore bottom-up edge control of graphene nanoribbons (GNRs) by functionalizing GNRs and related materials with various addends that lead to better performance in applications of interest to the AFOSR. The functionalization informs our understanding of the properties of GNRs at the edges and basal plane. The applications of interest to the AFOSR include anti-icing and active deicing of airfoils; new materials for energy storage and generation; and the use of functionalized GNRs in polymer composites. A method of using top-down laser lithography to produce GNR-like structures in laser induced graphene (LIG) was discovered in our lab, as disclosed in a prior annual report. We have explored the properties and uses of LIG in depth, as well as extremely short GNRs, called graphene quantum dots (GQDs), that can be synthesized from coal. GNRs, LIG and GQDs all have graphene and graphene-like edges that need to be understood and whose structures need to be controlled for obtaining the best properties for each application. AFOSR funding has been leveraged to partially fund the development of other materials, primarily for energy generation and storage applications.

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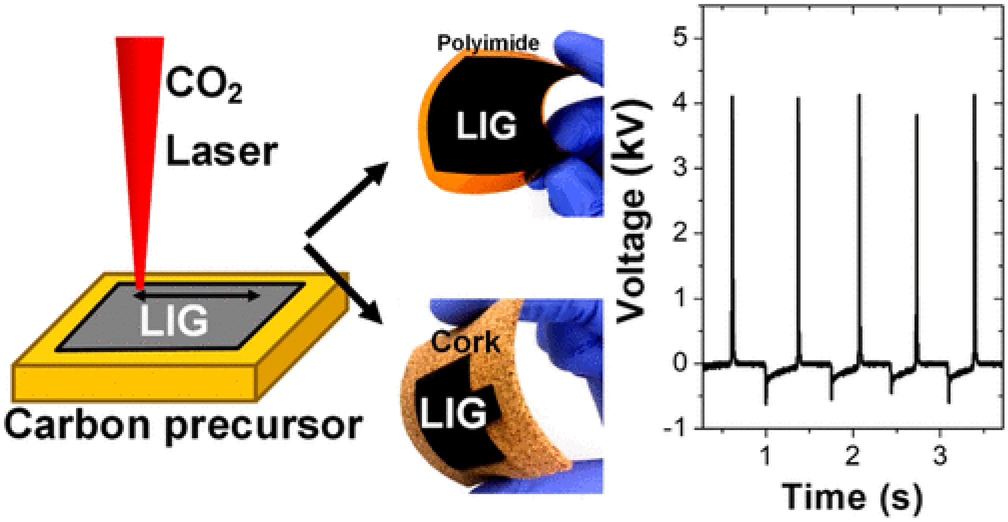
Abstract In this reporting period we have continued to explore bottom-up edge control of graphene nanoribbons (GNRs) by functionalizing GNRs and related materials with various addends that lead to better performance in applications of interest to the AFOSR. The functionalization informs our understanding of the properties of GNRs at the edges and basal plane. The applications of interest to the AFOSR include anti-icing and active deicing of airfoils; new materials for energy storage and generation; and the use of functionalized GNRs in polymer composites. A method of using top-down laser lithography to produce GNR-like structures in laser induced graphene (LIG) was discovered in our lab, as disclosed in a prior annual report. We have explored the properties and uses of LIG in depth, as well as extremely short GNRs, called graphene quantum dots (GQDs), that can be synthesized from coal. GNRs, LIG and GQDs all have graphene and graphene-like edges that need to be understood and whose structures need to be controlled for obtaining the best properties for each application. AFOSR funding has been leveraged to partially fund the development of other materials, primarily for energy generation and storage applications.

Research Fully or Partially Funded by the AFOSR:

Laser-Induced Graphene Triboelectric Nanogenerators

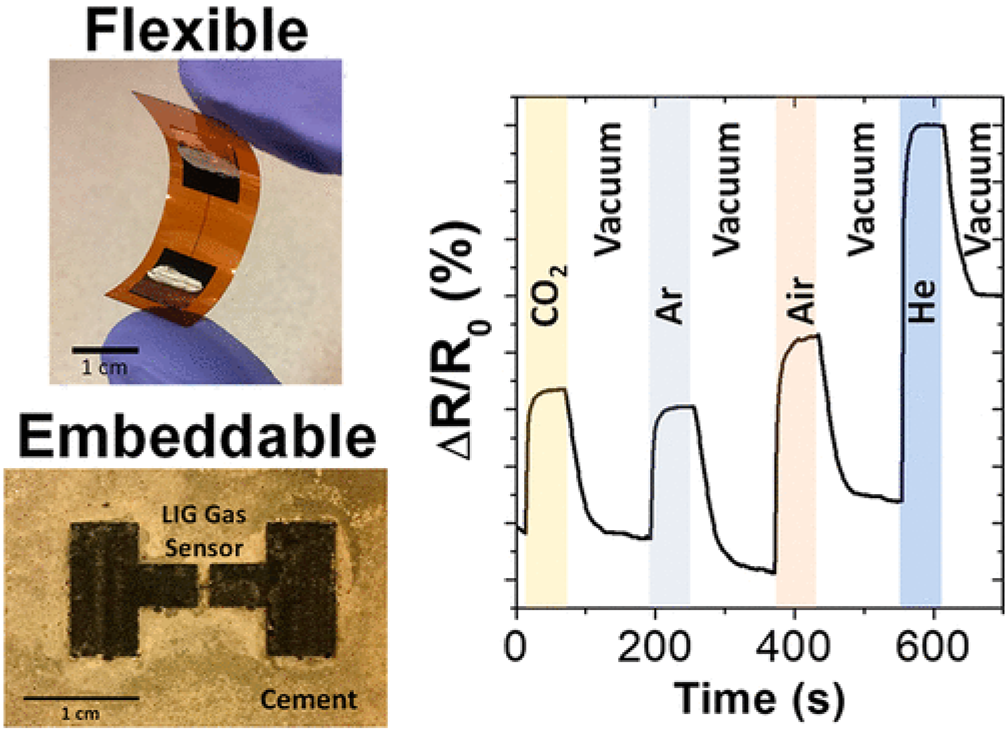
Executive Summary: Triboelectric nanogenerators (TENGs) show exceptional promise for converting wasted mechanical energy into electrical energy. This study investigates the use of laser-induced graphene (LIG) composites as an exciting class of triboelectric materials in TENGs. Infrared laser irradiation is used to convert the surfaces of the two carbon sources, polyimide (PI) and cork, into LIG. This gives the bilayer composite films the high conductivity associated with LIG and the triboelectric properties of the carbon source. A LIG/PI composite is used to fabricate TENGs based on conductor-to-dielectric and metal-free dielectric-to-dielectric device geometries with open-circuit voltages >3.5 kV and peak power >8 mW. Additionally, a single sheet of PI is converted to a metal-free foldable TENG. The LIG is also embedded within a PDMS matrix to form a single-electrode LIG/PDMS composite TENG. This single-electrode TENG is highly flexible and stretchable and was used to generate power from mechanical contact with skin. The LIG composites present a class of triboelectric materials that can be made from naturally occurring and synthetic carbon sources.



From: Stanford, M. G.; Li, J. T.; Chyan, Y.; Wang, Z.; Wang, W.; Tour, J. M. “Laser-Induced Graphene Triboelectric Nanogenerators,” *ACS Nano* **2019**, 13, 7166-7174. DOI: 10.1021/acsnano.9b02596

Laser-Induced Graphene for Flexible and Embeddable Gas Sensors

Executive Summary: Laser-induced graphene (LIG) has received much attention since it enables simple and rapid synthesis of porous graphene. This work presents a robust direct-write LIG-based gas sensor, which senses gases based on thermal conductivity, similar to a katharometer sensor. The gas sensors are fabricated by lasing polyimide substrates with a 10.6 μm CO_2 laser to synthesize LIG. This enables the formation of flexible gas sensors which could be incorporated on a variety of surfaces. High surface area and thermal conductivity of the LIG results in rapid response times for all studied gases. The gas sensors are also embedded in cement to form a refractory composite material. These sensors are used to determine composition of various gas mixtures, such as N_2 and CO_2, which are the most abundant gaseous species in flue gas. Thus, LIG based embeddable sensors could be incorporated in composites to enable electronically functional construction materials.



From: Stanford, M. G.; Yang, K.; Chyan, Y.; Kittrell, C.; Tour, J. M. "Laser-Induced Graphene for Flexible and Embeddable Gas Sensors," *ACS Nano* **2019** *13,* 3474-3482. DOI: 10.1021/acsnano.8b09622

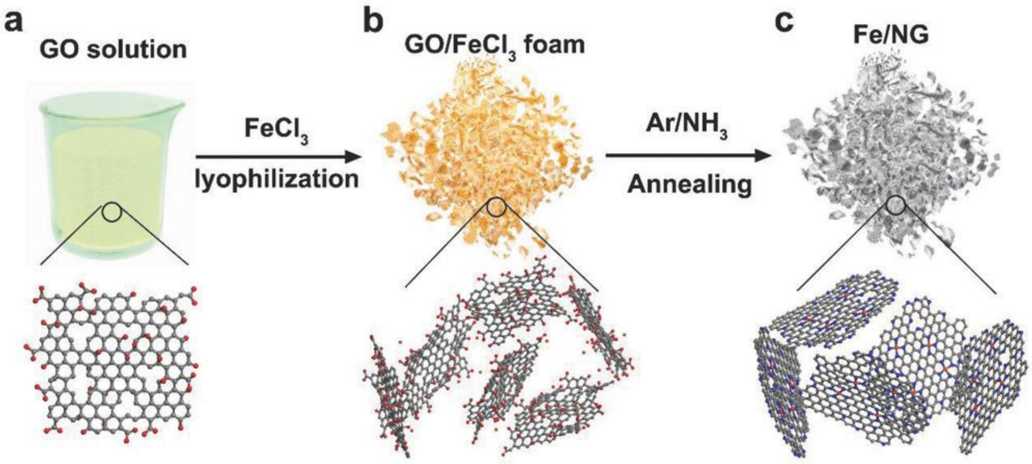
Achieving Self-Stiffening and Laser Healing by Interconnecting Graphene Oxide Sheets with Amine-Functionalized Ovalbumin

Executive Summary: Mimicking the remarkable properties of natural materials such as toughness, self-stiffening, self-healing, etc., is useful for several structural, functional, and biomedical applications. The judicious combination of natural and synthetic building blocks to design hybrid materials could offer an alternative route to achieving the aforementioned properties of natural materials. This study reports easily scalable process to synthesize graphene oxide (GO)/egg white hybrid material (polyalbumene) by cross-linking GO nanosheet with the amine-based cross-linker diethylenetriamine (DETA) and egg white. The result is a layered material like nacre, exhibiting high strength and toughness. Abundant functional groups on the GO allow covalent interactions between DETA and egg white protein amino acids. The atomistic simulations reveal that the insertion of GO into the composite has a sizeable impact on the composite's stiffness. As shown by experiments, the hybrid material has an elastic modulus of ≈41 MPa and a ductility of more than 25%. The hybrid material also exhibits a fracture repair property under laser exposure as well as CO_2 absorption and stiffening over time, suggesting an adaptable behavior. This work explores the possibility that the combination of synthetic and natural approaches offers a better route to designing advanced, tough, strong, adaptable, and fracture recovering materials.

From: Owuor, P. S.; Tsafack, T., Schara, S.; Hwang, H.; Jung, S.; Salvatierra, R. V.; Li, T.; Susarla, S.; Ren, M. Q.; Wei, B. Q.; Vajtai, R.; Tour, J. M.; Lou, J.; Tiwary, C. S.; Ajayan, P. M. "Achieving Self-Stiffening and Laser Healing by Interconnecting Graphene Oxide Sheets with Amine-Functionalized Ovalbumin," *Adv. Mater. Interf.* **2018**, *5*, 1800932. DOI: 10.1002/admi.201800932

Electrochemical CO_2 Reduction with Atomic Iron-Dispersed on Nitrogen-Doped Graphene

Executive Summary: Electrochemical reduction of CO_2 provides an opportunity to reach a carbon-neutral energy recycling regime, in which CO_2 emissions from fuel use are collected and converted back to fuels. The reduction of CO_2 to CO is the first step toward the synthesis of more complex carbon-based fuels and chemicals. Therefore, understanding this step is crucial for the development of high-performance electrocatalyst for CO_2 conversion to higher order products such as hydrocarbons. Here, atomic iron dispersed on nitrogen-doped graphene (Fe/NG) is synthesized as an efficient electrocatalyst for CO_2 reduction to CO. Fe/NG has a low reduction overpotential with high Faradic efficiency up to 80%. The existence of nitrogen-confined atomic Fe moieties on the nitrogen-doped graphene layer is confirmed by aberration-corrected high-angle annular dark-field scanning transmission electron microscopy and X-ray absorption fine structure analysis. The Fe/NG catalysts provide an ideal platform for comparative studies of the effect of the catalytic center on the electrocatalytic performance. The CO_2 reduction reaction mechanism on atomic Fe surrounded by four N atoms ($Fe–N_4$) embedded in nitrogen-doped graphene is further investigated through density functional theory calculations, revealing a possible promotional effect of nitrogen doping on graphene.

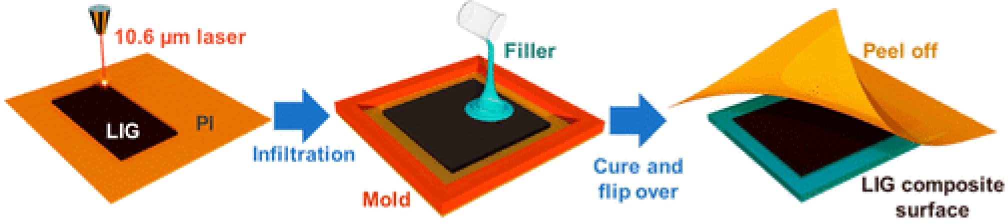


Schematic of the synthesis process of the Fe/NG catalyst. a) GO suspension in deionized water, b) Fe/NG precursor foam obtained from GO through a lyophilization process, and c) the final Fe/NG product obtained through the Ar/NH_3 annealing process at 650–800 °C.

From: Zhang, C.; Yang, S.; Wu, J.; Liu, M.; Yazdi, S.; Ren, M.; Sha, J.; Zhong, J.; Nie, K.; Jalilov, A. S.; Li, Z.; Li, H.; Yakobson, B. I.; Wu, Q.; Ringe, E.; Xu, H.; Ajayan, P. M.; Tour, J. M. "Electrochemical CO_2 Reduction with Atomic Iron-Dispersed on Nitrogen-Doped Graphene," *Adv. Energy Mater*. **2018**, *8*, 1703487. DOI: 10.1002/aenm.201703487

Laser-Induced Graphene Composites as Multifunctional Surfaces

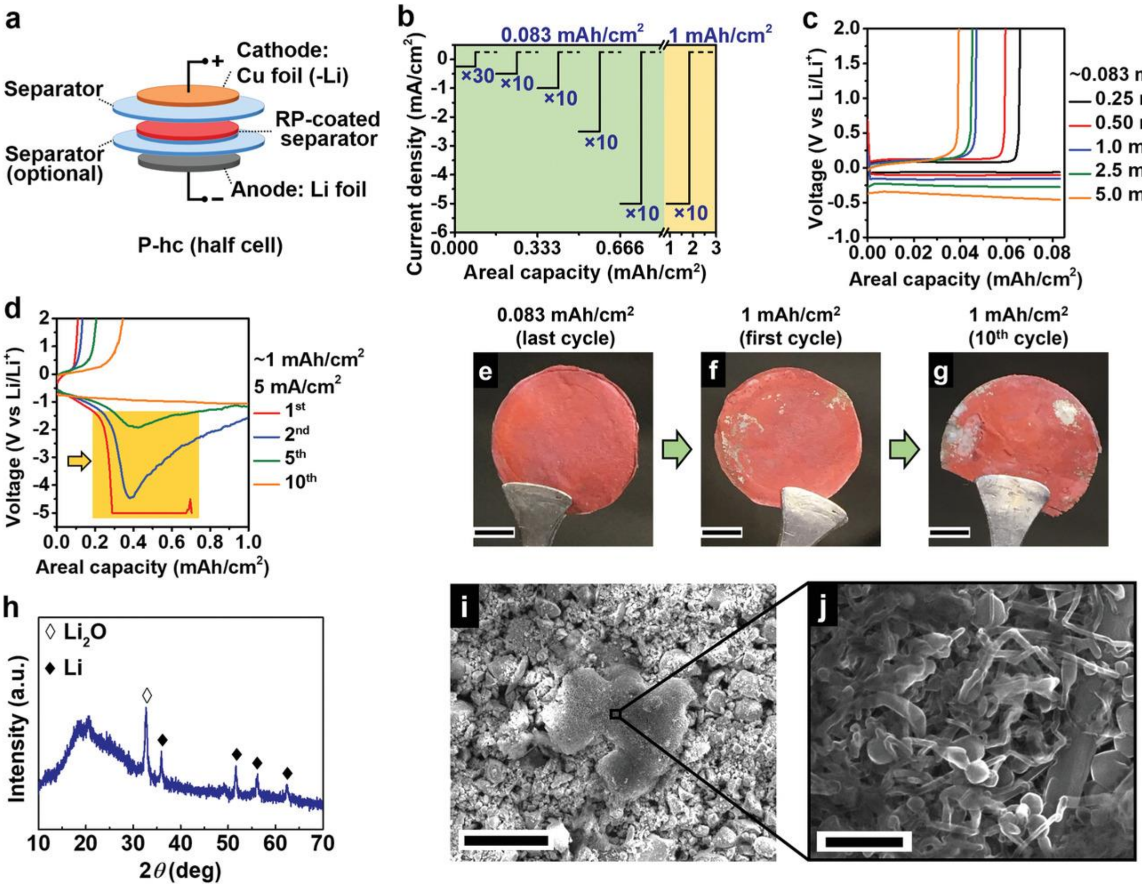
Executive Summary: Laser-induced graphene (LIG) is a platform material for numerous applications. Despite its ease in synthesis, LIG's potential for use in some applications is limited by its robustness on substrates. Here, using a simple infiltration method, we develop LIG composites (LIGCs) with physical properties that are engineered on various substrate materials. The physical properties include surface properties such as superhydrophobicity and antibiofouling; the LIGCs are useful in antibacterial applications and Joule-heating applications and as resistive memory device substrates.



From: Luong, D. X.; Yang, K.; Yoon, J.; Singh, S. P.; Wang, T.; Arnusch, C. J.; Tour, J. M. "Laser-Induced Graphene Composites as Multifunctional Surfaces," *ACS Nano* **2019**, *13*, 2579-2586. DOI: 10.1021/acsnano.8b09626

Detecting Li Dendrites in a Two-Electrode Battery System

Executive Summary: The use of high-energy-density Li metal anodes in rechargeable batteries is not possible because of dendrite formation that can potentially result in a battery fire. Although so-called dendrite-free Li metal anodes have been reported in many recent publications, Li dendrite growth is still kinetically favorable and it remains a severe safety concern in mass production. Here, a detection system capable of alerting for Li dendrite formation in a two-electrode battery with no additional electrodes required is reported. When dendrites contact a red phosphorous-coated separator, dendrite growth is revealed by a significant voltage change. This can activate a signal through the battery management system, warning of the presence of Li dendrites and leading to shutdown of the battery before the dendrites become dangerous.

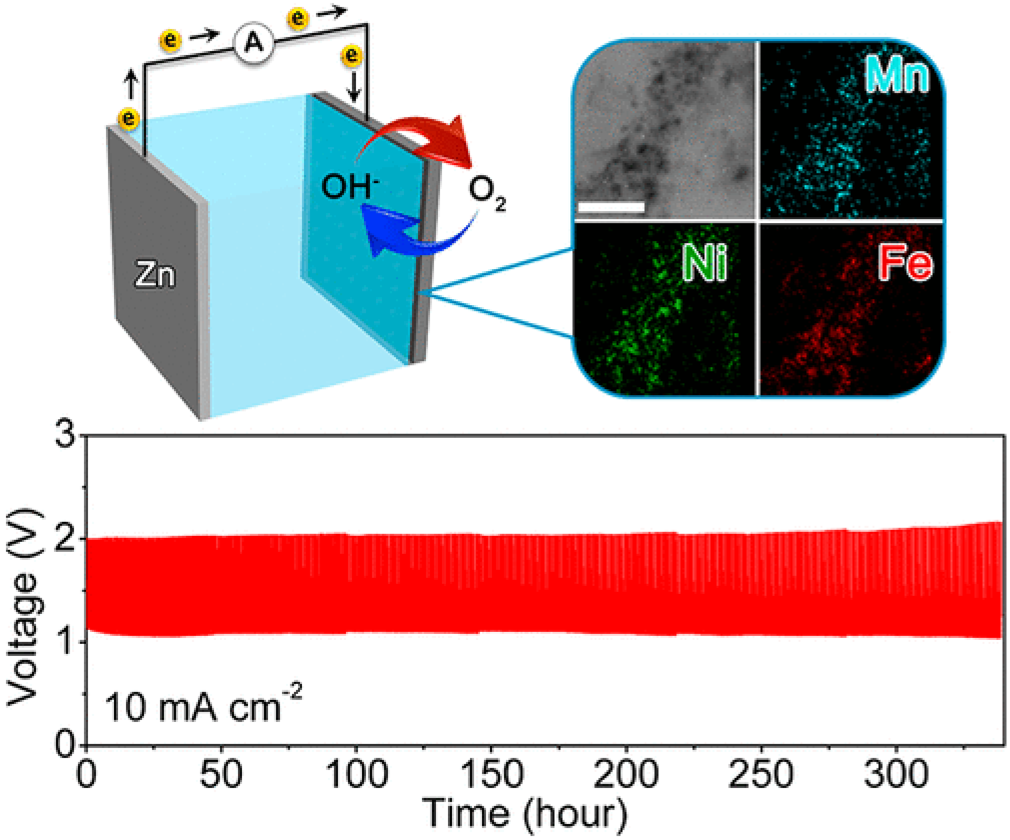


Half-cell tests with the red phosphorous (RP)-coated separator. a) Configuration of the half-cell with RP coating (P-hc). b) Testing protocol showing the current density versus time. The discharge steps were time-control (20/10/5/2/1 min at ≈0.083 mAh cm^{-2}and 12 min at ≈1 mAh cm^{-2}); the charge steps were voltage-control (2 V) and are thus shown as dashed lines because the areal capacities (<0.083 mAh cm^{-2}) varied. Voltage profiles at c) ≈0.083 mAh cm^{-2}and d) ≈1 mAh cm^{-2}. The voltage decrease indicated by the arrow is the electrochemical response generated when Li dendrites contact the RP. e–g) Photographs of the RP-coated separator at different stages showing e) no silver-colored Li metal and f,g) Li metal on the RP. h) XRD spectrum of the silver-colored spots in g). i,j) SEM images of the silver-colored spots in g showing the dendritic morphology of Li. Scale bars: i) 100 µm; j) 2 µm.

From: Wang, T.; Salvatierra, R. V.; Tour, J. M. Detecting Li Dendrites in a Two-Electrode Battery System,” *Adv. Mater.* **2019** 1807405. DOI: 10.1002/adma.201807405

Laser-Induced Graphene Hybrid Catalysts for Rechargeable Zn-Air Batteries

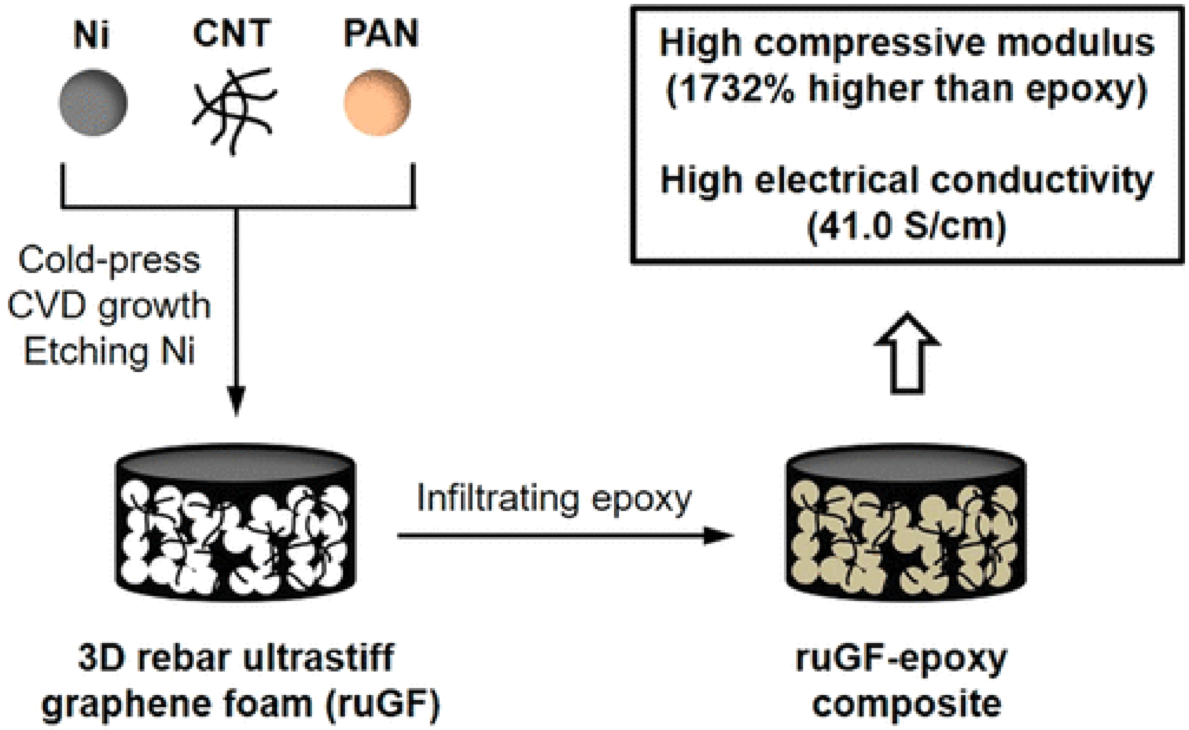
Executive Summary: Rechargeable Zn-air batteries are projected to afford electrical energy storage with high energy and power density, safety, and economic viability. One of the key components in designing the battery is the cathode catalyst that is used to facilitate the cathodic reactions, specifically oxygen reduction and oxygen evolution reactions (ORR and OER). Here we report the facile synthesis of ternary metal oxide/graphene hybrid catalysts by combining ORR-active Co/Mn with OER-active Ni and Fe species to promote the bifunctional activity all in an in situ formed laser-induced graphene flexible film. These hybrid catalysts exhibit high catalytic activity and surpass the performance of precious metal Pt and RuO_2 catalysts in Zn-air batteries. The batteries show high discharge peak power density of 98.9 mW cm^{-2} and energy density of 842 Wh kg_{Zn}^{-1}. The batteries also show high reversibility and durability through charge/discharge cycles for >200 h. The catalysts demonstrate that applications in flexible Zn-air batteries that would be beneficial for wearable and flexible electronic devices.



From: Ren, M; Zhang, J.; Tour, J. M. “Laser-Induced Graphene Hybrid Catalysts for Rechargeable Zn-Air Batteries,” *ACS Appl. Energy Mater.* **2019**, *2*, 1460-1468. DOI: 10.1021/acsaem.8b02011

Ultra-Stiff Graphene Foams as Three-Dimensional Conductive Fillers for Epoxy Resin

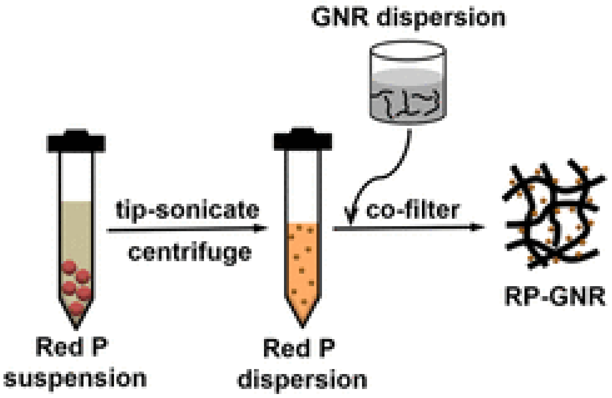
Executive Summary: Conductive epoxy composites are of great interest due to their applications in electronics. They are usually made by mixing powdered conductive fillers with epoxy. However, the conductivity of the composite is limited by the low filler content because increasing filler content causes processing difficulties and reduces the mechanical properties of the epoxy host. We describe here the use of ultra-stiff graphene foams (uGFs) as three-dimensional (3D) continuous conductive fillers for epoxy resins. The powder metallurgy method was used to produce the dense uGFs monoliths that resulted in a very high filler content of 32 wt % in the uGF–epoxy composite, while the density of epoxy was only increased by 0.09 g/cm^3. The composite had an electrical conductivity of 41.0 ± 6.3 S/cm, which is among the highest of all of the polymer-based composites with non-conductive polymer matrices and comparable with the conductive polymer matrices reported to date. The compressive modulus of the composite showed a remarkable improvement of >1700% compared to pure epoxy. We have demonstrated that the 3D uGF filler substantially improves the conductivity and reinforces the polymer matrix with a high filler content while retaining a density similar to that of the epoxy alone.

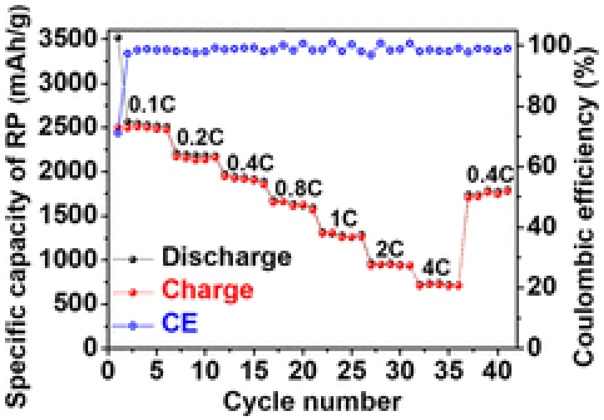


From: Han, X.; Wang, T.; Owuor, P. S.; Hwang, S. H.; Cao Wang, C.; Sha, S.; Shen, L.; Yoon, J.; Wang, W.; Salvatierra, R. V.; Ajayan, P. M.; Shahsavari, R.; Lou, J.; Zhao, Y.; Tour, J. M. “Ultra-Stiff Graphene Foams as Three-Dimensional Conductive Fillers for Epoxy Resin,” *ACS Nano* **2018**, 12, 11219-11228. DOI: 10.1021/acsnano.8b05822

Tip-Sonicated Red Phosphorus-Graphene Nanoribbon Composite for Full Lithium-Ion Batteries

Executive Summary: Red phosphorus (RP) is considered a promising anode material for lithium-ion batteries (LIBs) due to its high energy density and low cost. Although RP is electrically insulating, researchers have reduced its particle size and added conductive fillers to improve the electrochemical activity of RP. Here, we report a method for making <1 µm sized RP under ambient conditions by using tip sonication. A specific surfactant solution was used to stabilize the dispersion of <1 µm sized RP. Graphene nanoribbons (GNRs) were added to improve the conductivity. The RP–GNR composite achieved nearly maximum capacity at 0.1C and showed a capacity retention of 96% after 216 cycles at 0.4 C in the half-cell. When combined with a $LiCoO_2$ cathode, the full cell delivered a total capacity of 86 mAh/g after 200 cycles at 0.4C. This study has demonstrated the fabrication of high-performance LIBs using RP in a safe, convenient, and cost-effective manner, and the method might be extended for the preparation of other battery or catalyst materials that are difficult to acquire through bottom-up or top-down approaches.

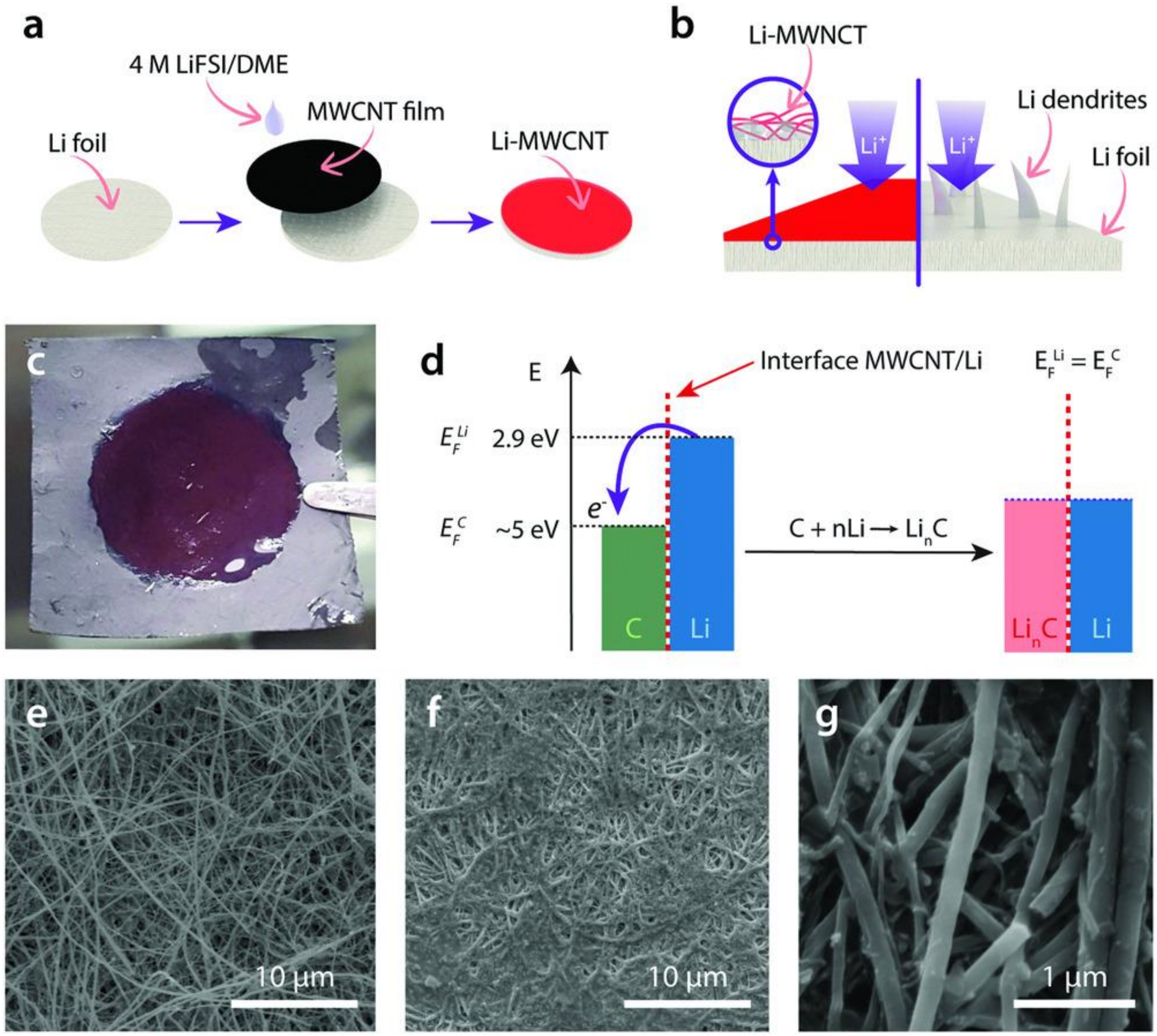




From: Wang, T.; Wei, S.; Salvatierra, R. V.; Han, X.; Wang, Z.; Tour, J. M. "Tip-Sonicated Red Phosphorus-Graphene Nanoribbon Composite for Full Lithium-Ion Batteries," *ACS Appl. Mater. Interfaces* **2018**, *10*, 38936-38943. DOI: 10.1021/acsami.8b14073

Suppressing Li Metal Dendrites Through a Solid Li-Ion Backup Layer

The growing demand for sustainable and off-grid energy storage is reviving the attempts to use Li metal as the anode in the next generation of batteries. However, the use of Li anodes is hampered due to the growth of Li dendrites upon charging and discharging, which compromises the life and safety of the battery. Here, it is shown that lithiated multiwall carbon nanotubes (Li-MWCNTs) act as a controlled Li diffusion interface that suppresses the growth of Li dendrites by regulating the Li^+ ion flux during charge/discharge cycling at current densities between 2 and 4 mA cm^{-2}. A full Li-S cell is fabricated to showcase the versatility of the protected Li anode with the Li-MWCNT interface, where the full cells could support pulse discharges at high currents and over 450 cycles at different rates with coulombic efficiencies close to 99.9%. This work indicates that carbon materials in lithiated forms can be an effective and simple approach to the stabilization of Li metal anodes.

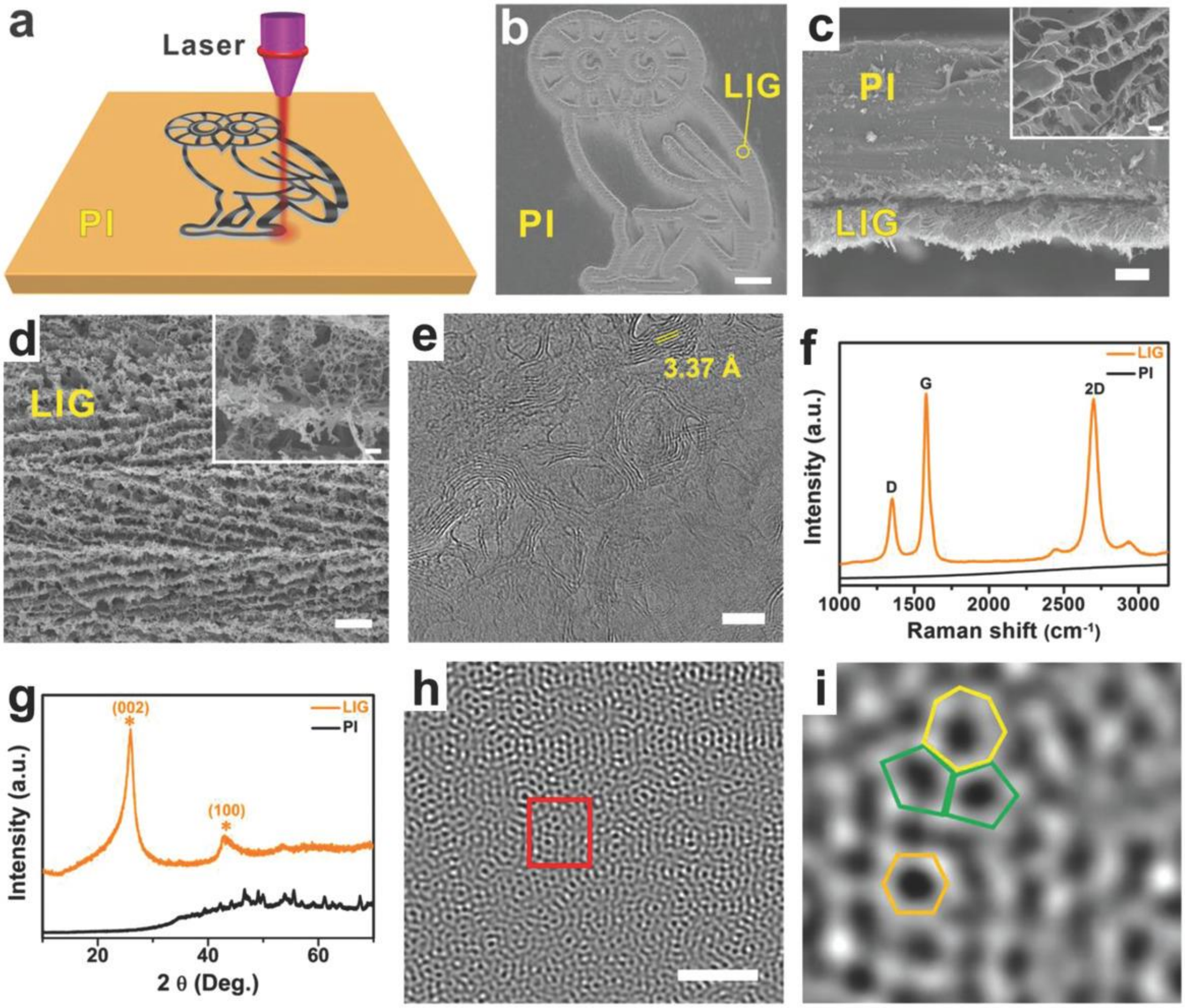


Schematic of the Li metal protected by the Li-MWCNT interface, its fabrication, and morphology characterization. The final structure of the electrode is composed of Li metal coated with Li-MWCNT. a) The fabrication process of the Li-MWCNT electrode consisted of wetting a MWCNT film with a highly concentrated electrolyte and pressing it against a Li foil. b) Li-MWCNT serves as a Li^+ ion source on demand that controls the Li plating/stripping (left). This mediation process suppresses the dendrite growth that occurs on the Li metal surface (right). c) Photograph of MWCNT film after being lithiated; the lithiated area is red. d) Scheme of the spontaneous lithiation of MWCNTs and the corresponding redox reaction. Energy diagrams demonstrate the driving force for reduction of MWCNTs based on the difference of Fermi energy levels (work function) of Li (−2.9 eV versus vacuum) and C (−5 eV versus vacuum). SEM images of e) the pristine MWCNT film and f,g) the Li-MWCNT film.

From: Salvatierra, R. V.; López-Silva, G. A.; Jalilov, A. S.; Yoon, J.; Wu, G.; Tsai, A.-L.; Tour, J. M. "Suppressing Li Metal Dendrites Through a Solid Li-Ion Backup Layer," *Adv. Mater*. **2018**, 1803869. DOI: 10.1002/adma.201803869

Laser-Induced Graphene: From Discovery to Translation (a review)

Executive Summary: Laser-induced graphene (LIG) is a 3D porous material prepared by direct laser writing with a CO_2 laser on carbon materials in ambient atmosphere. This technique combines 3D graphene preparation and patterning into a single step without the need for wet chemical steps. Since its discovery in 2014, LIG has attracted broad research interest, with several papers being published per month using this approach. These serve to delineate the mechanism of the LIG-forming process and to showcase the translation into many application areas. Herein, the strategies that have been developed to synthesize LIG are summarized, including the control of LIG properties such as porosity, composition, and surface characteristics, and the advancement in methodology to convert diverse carbon precursors into LIG. Taking advantage of the LIG properties, the applications of LIG in broad fields, such as microfluidics, sensors, and electrocatalysts, are highlighted. Finally, future development in biodegradable and biocompatible materials is briefly discussed.



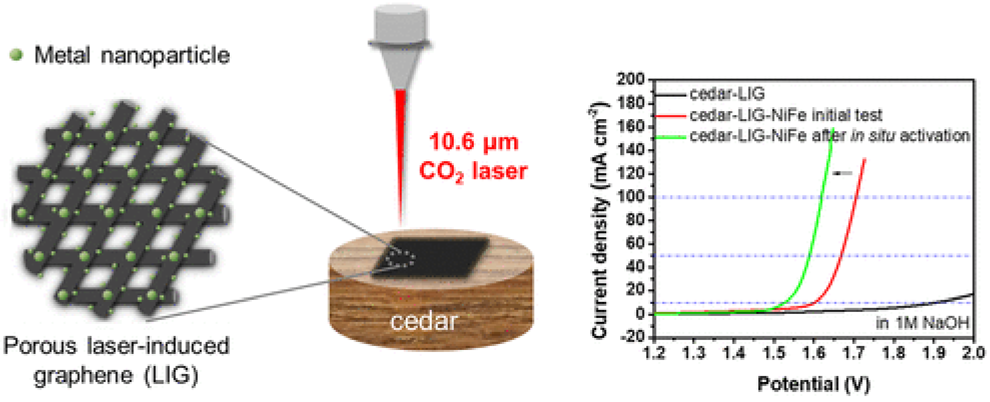
LIG formation on PI. a) Schematic of the synthesis process of LIG from PI using a CO_2 laser. b) SEM image of LIG patterned into an owl shape; scale bar is 1 mm. The bright contrast corresponds to LIG surrounded by the darker-colored insulating PI substrate. c) Cross-sectional

SEM image of the LIG film on the PI substrate; scale bar, 20 µm. Inset is the SEM image showing the porous morphology of LIG; scale bar, 1 µm. d) SEM image of the LIG film circled in (b); scale bar is 10 µm. Inset is the corresponding higher magnification SEM image; scale bar is 1 µm. e) HRTEM image of LIG; scale bar is 5 nm. Average lattice space of ≈3.4 Å corresponds to the (002) planes of graphitic materials. f) Representative Raman spectrum of LIG film and the starting PI film. g) XRD of powdered LIG scraped from the PI film showing turbostratic layering. h) Cs-correction STEM image taken at the edge of an LIG flake; scale bar, 2 nm, showing an ultra-polycrystalline nature with grain boundaries. i) TEM image of selected area indicated as a rectangle in (h)). It shows a heptagon with two pentagons as well as a hexagon. a–i).

From: Ye, R.; James, D. K.; Tour, J. M. “Laser-Induced Graphene. From Discovery to Translation,” *Adv. Mater*. **2018**, 1803621, DOI: 10.1002/adma.201803621.

Laser-Induced Graphene from Wood Impregnated with Metal Salts and Use in Electrocatalysis

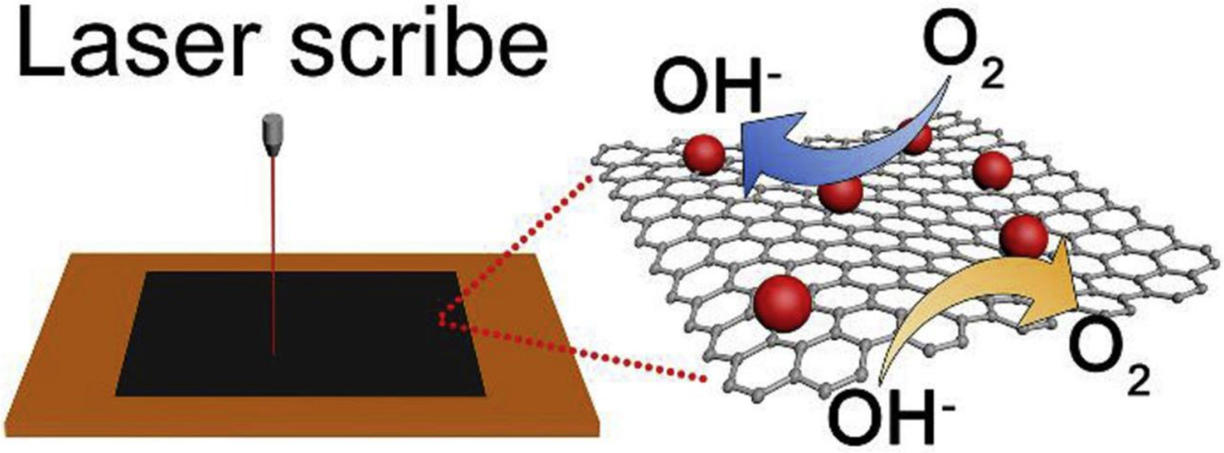
Executive Summary: The hybridization of graphene with other inorganic nanostructures has endowed graphene with enhanced and varied functionalities. Here we demonstrate a facile and improved approach to convert biodegradable cedar wood into graphene embedded with various metal nanocrystals (cedar-LIG-M, LIG is laser-induced graphene, M = Cu, Co, Ni, Fe, NiFe) by sonication-assisted soaking and one-step CO_2 laser scribing. Organic biomass was transformed to hierarchical porous graphene via laser induction, whereas metal salts were reduced to elemental metals simultaneously by the carbothermal reaction and reducing atmosphere generated during the lignocellulose decomposition. The as-prepared cedar-LIG-M possesses an ordered porous structure, good conductivity, unique ferromagnetic behavior and excellent electrochemical catalytic performance. As a demonstration, the cedar-LIG-NiFe electrode has a low overpotential of 296 mV at a current density of 10 mA cm^{-2}for oxygen evolution reactions. The performance of the electrode continued to improve at the initial testing stage due to the in situ activation as a result of the increased oxidation states of nickel and iron during electrochemical oxygen evolution. In addition, the cedar-LIG-NiFe could also serve as an electromagnetic interference shielding material with shielding effectiveness up to 54 dB. The simplicity and versatility of this technique provides a route for the synthesis of various carbon-based hybrid materials with potential applications of the products in many different fields, such as energy storage, electrocatalysis, electromagnetic interference shielding, and water treatment.



From: Han, X.; Ye, R.; Chyan, Y.; Wang, T.; Zhang, C.; Shi, L.; Zhang, T.; Zhao, Y.; Tour, J. M. "Laser-Induced Graphene from Wood Impregnated with Metal Salts and Use in Electrocatalysis," *ACS Appl. Nano Mater.*, **2018**, *9*, 5053-5061. DOI: 10.1021/acsanm.8b01163

Laser-induced graphene synthesis of Co_3O_4 in graphene for oxygen electrocatalysis and metal-air batteries

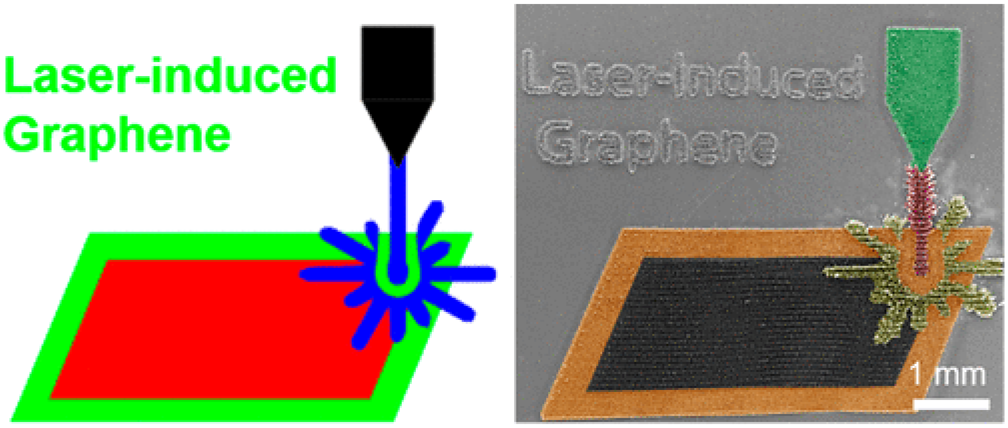
Executive Summary: We present here a straightforward synthesis of highly efficient bifunctional OER/ORR catalysts through a facile laser-induced graphene (LIG) process to produce Co_3O_4/LIG. The Co_3O_4/LIG showed OER and ORR activity comparable to noble metal-based catalysts in alkaline electrolyte. Furthermore, the Co_3O_4/LIG exhibited promising performance in Zn-air and Li-O_2 batteries. The rechargeable Zn-air battery has an open-circuit potential of 1.46 V and a high power density of 84.2 mW/cm^2 at 100 mA/cm^2. The Li-O_2 battery with the Co_3O_4/LIG cathode exhibits low overpotentials in both charge and discharge processes and excellent cycling stability up to 242 cycles.



From: Ren, M.; Zhang, J.; Tour, J. M. "Laser-Induced Graphene Synthesis of Co_3O_4 in Graphene for Oxygen Electrocatalysis and Metal-Air Batteries," *Carbon* **2018**, *139*, 880-887. https://doi.org/10.1016/j.carbon.2018.07.051

Laser-Induced Graphene (a review)

Executive Summary: Research on graphene abounds, from fundamental science to device applications. In pursuit of complementary morphologies, formation of graphene foams is often preferred over the native two-dimensional (2D) forms due to the higher available area. Graphene foams have been successfully prepared by several routes including chemical vapor deposition (CVD) methods and by wet-chemical approaches. For these methods, one often needs either high temperature furnaces and highly pure gases or large amounts of strong acids and oxidants. In 2014, using a commercial laser scribing system as found in most machine shops, a direct lasing of polyimide (PI) plastic films in the air converted the PI into 3D porous graphene, a material termed laser-induced graphene (LIG). This is a one-step method without the need for high-temperature reaction conditions, solvent, or subsequent treatments, and it affords graphene with many five-and seven-membered rings. With such an atomic arrangement, one might call LIG "kinetic graphene" since there is no annealing in the process that causes the rearrangement to the preferred all-six-membered-ring form. In this Account, we will first introduce the approaches that have been developed for making LIG and to control the morphology as either porous sheets or fibrils, and to control porosity, composition, and surface properties. The surfaces can be varied from being either superhydrophilic with a 0° contact angle with water to being superhydrophobic having >150° contact angle with water. While it was initially thought that the LIG process could only be performed on PI, it was later shown that a host of other polymeric substrates, nonpolymers, metal/plastic composites, and biodegradable and naturally occurring materials and foods could be used as platforms for generating LIG. Methods of preparation include roll-to-roll production for fabrication of in-plane electronics and two different 3D printing (additive manufacturing) routes to specific shapes of LIG monoliths using both laminated object manufacturing and powder bed fabrication methods. Use of the LIG in devices is performed very simply. This is showcased with high performance supercapacitors, fuel cell materials for oxygen reduction reactions, water splitting for both hydrogen and oxygen evolution reactions coming from the same plastic sheet, sensor devices, oil/water purification platforms, and finally applications in both passive and active biofilm inhibitors. So the ease of formation of LIG, its simple scale-up, and its utility for a range of applications highlights the easy transition of this substrate-bound graphene foam into commercial device platforms.



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