

Graphitic Design: Prospects of Graphene-Based Nanocomposites for Solar Energy Conversion, Storage, and Sensing

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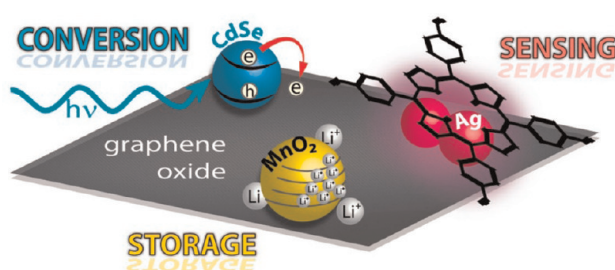
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CONSPECTUS

Graphene not only possesses interesting electrochemical behavior but also has a remarkable surface area and mechanical strength and is naturally abundant, all advantageous properties for the design of tailored composite materials. Graphene–semiconductor or –metal nanoparticle composites have the potential to function as efficient, multifunctional materials for energy conversion and storage. These next-generation composite systems could possess the capability to integrate conversion and storage of solar energy, detection, and selective destruction of trace environmental contaminants or achieve single-substrate, multistep heterogeneous catalysis. These advanced materials may soon become a reality, based on encouraging results in the key areas of energy conversion and sensing using graphene oxide as a support structure. Through recent advances, chemists can now integrate such processes on a single substrate while using synthetic designs that combine simplicity with a high degree of structural and composition selectivity. This progress represents the beginning of a transformative movement leveraging the advancements of single-purpose chemistry toward the creation of composites designed to address whole-process applications.

The promising field of graphene nanocomposites for sensing and energy applications is based on fundamental studies that explain the electronic interactions between semiconductor or metal nanoparticles and graphene. In particular, reduced graphene oxide is a suitable composite substrate because of its two-dimensional structure, outstanding surface area, and electrical conductivity. In this Account, we describe common assembly methods for graphene composite materials and examine key studies that characterize its excited state interactions. We also discuss strategies to develop graphene composites and control electron capture and transport through the 2D carbon network. In addition, we provide a brief overview of advances in sensing, energy conversion, and storage applications that incorporate graphene-based composites. With these results in mind, we can envision a new class of semiconductor– or metal–graphene composites sensibly tailored to address the pressing need for advanced energy conversion and storage devices.



The favorable electronic properties of graphene, in addition to its impressive surface area, mechanical strength, and natural abundance, offer new opportunities for the design of composite materials with tailored properties.^{1–5} Chemical exfoliation of graphene to produce graphene oxide (GO) is a convenient route to synthesize large batches of the single-layer material, which readily suspends in polar solvents.^{6–8} Postsynthesis, GO can be further modified with semiconductor and metal nanoparticles.^{9–11} Functional groups such as –OH, –COOH, and epoxide along with

structural defects serve as anchoring points for introducing selective functionality including electron acceptor or donor groups.^{12,13} Additionally, the two-dimensional sp² carbon network facilitates direct interaction with sheet-like inorganic structures and planar organic molecules.¹⁴ For example, porphyrin molecules interact via π – π interactions making the assembly photoactive.^{15,16} Both suspensions and films of chemically exfoliated GO can be easily reduced to partially restore the extended sp² carbon–carbon network found in pristine graphene.^{17–20} Following the

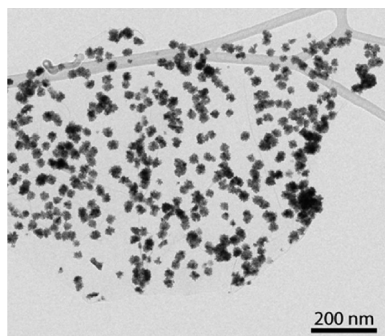


FIGURE 1. Platinum nanoparticles deposited on RGO. A dual frequency method reduces GO and metal ions while ensuring GO is continually exfoliated. Reprinted with permission from ref 36. Copyright 2012 Elsevier.

reduction process, the material is referred to as reduced graphene oxide (RGO).

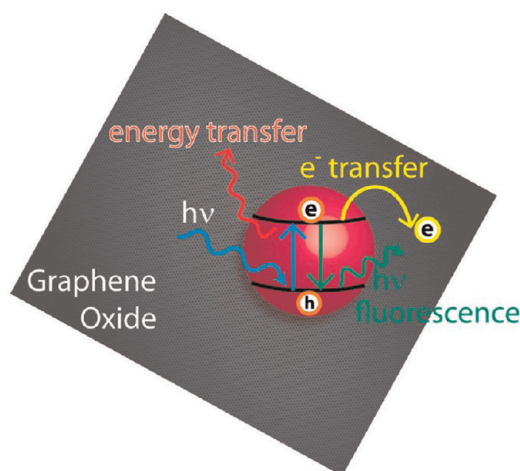
The 2D carbon mat of graphene-based assemblies has found use in a variety of interesting applications. For example, RGO is incorporated in plastic composites to create stronger conducting plastics and has also been employed as a transparent conducting material in all-organic LEDs.^{21,22} Additionally, RGO is used as a delivery medium for cancer-fighting drugs and is combined with polyaniline nanofibers and semiconductors to make supercapacitor materials.^{23–25} Pertinent to this Account, RGO composites have found useful application in catalysis, energy conversion and storage, and trace molecule sensing.^{4,26,27} Design strategies for the development of graphene composites and manipulation of the electron capture and transport through the 2D-carbon network are discussed in this Account.

Designing Reduced Graphene Oxide–Metal or –Semiconductor Composites

Since GO or RGO acts as a stabilizer, GO–metal composites are prepared in the absence of additional stabilizers or surfactants.^{28,29} One of the early demonstrations of GO films as a stabilizer for silver nanoparticles (NPs) came from Fendler's group in the 1990s.³⁰ A number of deposition techniques have been reported in recent years with the aim of selective size control of catalytically active metals such as Ag, Au, Pt, and Pd.^{19,31–34} Chemical reductants such as hydrazine and NaBH₄ induce simultaneous reduction of metal ions and graphene oxide resulting in RGO–metal nanoparticle (NP) composites.

Microwave and sonolysis are also useful in designing metal–RGO composites.^{35,36} For example, sonication of a metal ion–GO dispersion in the presence of poly(ethylene glycol) results in high-coverage metal nanoparticle–RGO

SCHEME 1. Semiconductor Nanoparticles under Band Gap Illumination Are Capable of Reducing GO via Photogenerated Electron Transfer; Other Relaxation Pathways Include Energy Transfer and Recombination Resulting in Fluorescence



sheets.³⁷ An additional benefit of sonication-induced reduction is the active exfoliation of individual RGO sheets facilitated by sonication. This approach leads to metal NP deposition on single-layer RGO sheets with good dispersion. Figure 1 shows Pt–RGO composites formed using dual-frequency sonication. The method uses a high sonolytic frequency (211 kHz), which is effective for co-reducing the metal ions and GO, in conjunction with a low frequency (20 kHz), which is useful for maintaining GO sheet exfoliation.³⁶ Other methods for inducing metal NP deposition on graphene include co-reduction of GO and metal ions using microwave irradiation or gentle heating in the presence of ethylene glycol.^{38,39}

Photocatalytic deposition of metal NPs is another technique used for the formation of graphene–metal NP composites.²⁷ Photodeposition is accomplished by pairing a semiconductor with GO, making the material photoactive. In the presence of metal ions, illumination of the semiconductor–GO composite results in metal NP deposition on graphene following electron transfer from semiconductor to RGO to metal. Electron transfer in this process also results in reduction of GO.

Dispersions of physically mixed GO–semiconductor NPs provide a convenient route to form composite films on conducting electrode surfaces. Greater interaction and dispersion of semiconducting NPs on GO can be achieved by directly linking semiconductor NPs to GO via selectivity of functional groups. For example, the binding of TiO₂ NPs is augmented by the presence of –COOH groups of GO.⁴⁰ The pathways available for excited state electron relaxation following band gap excitation of a semiconductor NP anchored to graphene are depicted in Scheme 1. Electron

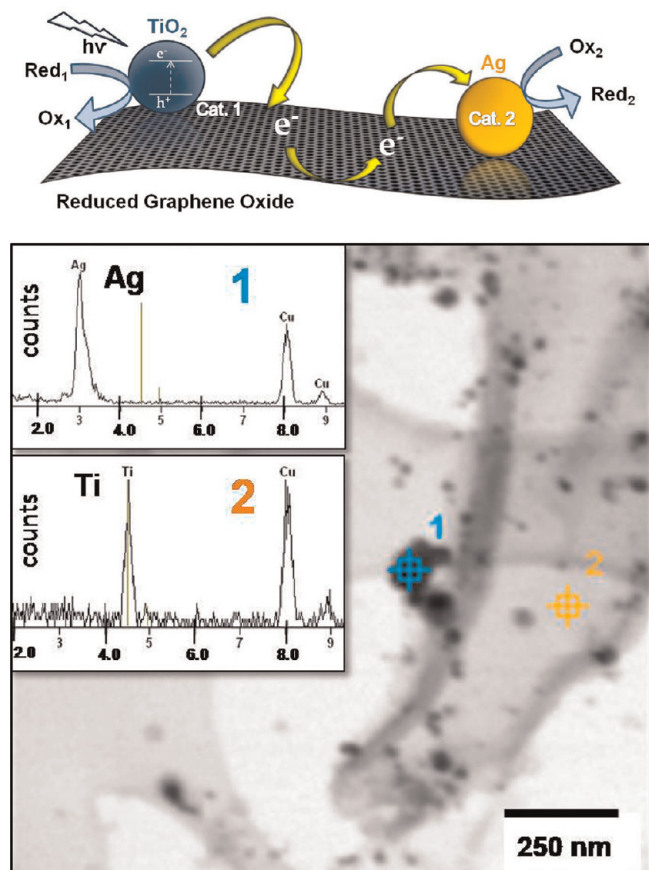


FIGURE 2. (top) Schematic illustrating electron transfer from photoexcited TiO₂ to RGO where it is shuttled to another site to reduce Ag⁺ ions to Ag NPs. (bottom) Scanning transmission electron micrograph showing distinct locations for TiO₂ and Ag on RGO. Adapted with permission from ref 27. Copyright 2010 American Chemical Society.

transfer resulting from band gap excitation of the semiconductor particles provides a straightforward technique to reduce GO.²⁷ UV-illuminated ZnO and TiO₂ have been investigated in detail to establish the transfer of photogenerated electrons to GO.^{20,41}

Electron Storage and Shuttling in RGO

An important quality of RGO is its ability to capture and shuttle electrons through its sp² network. Electrons captured by RGO from radicals or excited semiconductor NPs can be utilized to reduce metal ions. For example, the role of RGO as an electron shuttle is confirmed by the elucidation of charge transfer between molecular species and RGO. The photogenerated radical species of methyl viologen, MV^{•+}, transfers electrons to GO in a manner similar to photoexcited TiO₂ NPs.⁴²

The versatility of RGO as an electron shuttle is further confirmed by the quantitative elucidation of charge transfer between RGO and excited semiconductor NPs. In this

context, a quantitative electron titration methodology has recently been developed (Figure 2, top).²⁷ The controlled electron transfer process is initiated using UV-illumination to generate a finite bank of stored electrons in a N₂-purged, colloidal TiO₂ solution in ethanol. Under these conditions, it is possible to store and quantify electrons in surface defect trap sites on the TiO₂.⁴³ These electrons can then be titrated from the TiO₂ by incremental addition of a N₂-purged GO dispersion in which GO serves as the electron acceptor and is consequently reduced to RGO. Additional transferred electrons not consumed in GO's reduction are stored in RGO. Storage of electrons in RGO is similar to the electron accepting and storage capabilities seen in other carbon allotropes (viz., fullerenes and carbon nanotubes).^{43,44} Stored electrons in RGO are readily transferred to another electron-accepting species when added to the solution (e.g., Ag⁺ ions). The quantity of metal ions reduced by electrons stored in RGO can be estimated by tracking the growth in absorbance corresponding to the metal's surface plasmon resonance following each sequential addition of Ag⁺ ions. The storage and shuttling of electrons is realized from the deposition of silver particles at sites distinct from TiO₂ NPs (Figure 2). The ability to anchor semiconductor and metal NPs at different sites on RGO is useful for designing site-selective catalytic processes. Additionally, it offers opportunities for sensing through the combination of RGO with semiconductors, metal NPs, or other redox-active molecular species such as porphyrins.

Excited State Interactions

In addition to semiconductor and metal NPs, charge transfer interactions between RGO and photoactive molecular species such as porphyrins have been recently reported.^{15,16,42} For example, strong interactions between RGO and 5,10,15,20-tetrakis(1-methyl-4-pyridinio)porphyrin tetra(*p*-toluenesulfonate), or TMPyP, is evidenced by a red shift in porphyrin absorbance.¹⁴ The interactions in the excited state can be followed through fluorescence quenching, diminished excited state lifetimes, and monitoring the formation of electron transfer products. The formation of the TMPyP cation radical observed following laser pulse excitation of TMPyP-bound RGO confirms electron transfer in such composites.¹⁶ Several applications, including biosensing, nonlinear optics, and the creation of conducting films, take advantage of the electronic interactions observed in RGO–porphyrin composites.^{15,45,46}

The nature of excited state interactions between UV-illuminated semiconductor NPs (e.g., CdSe and ZnO) and RGO can be readily probed using fluorescence and

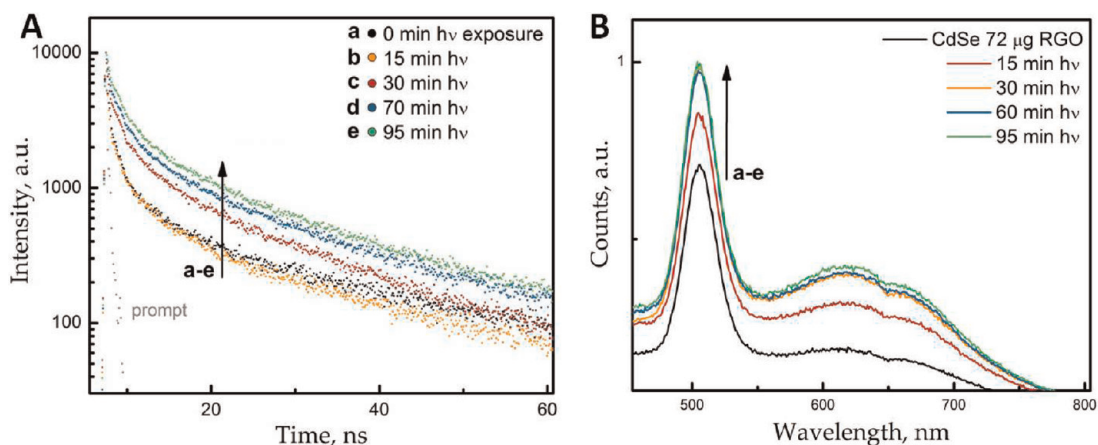


FIGURE 3. (A) CdSe–GO composites show increased photoluminescence lifetimes upon visible light irradiation (>420 nm) as charging of RGO inhibits electron transfer. (B) Emission recovery of CdSe–RGO solution upon illumination with visible light (>420 nm), relative to CdSe only emission. Adapted with permission from ref 49. Copyright 2012 American Chemical Society.

fluorescence lifetime decay analysis.^{47–49} The green emission of ZnO colloidal particles arising from the oxygen vacancy is quenched by RGO.⁴¹ The lifetime analysis indicates an electron transfer rate constant of $1.2 \times 10^9 \text{ s}^{-1}$ in ZnO–RGO composites. The reduction of GO is also reflected in increased conductivity of films following the photocatalytic reduction of GO.²⁰

In addition to electron transfer, there is also a possibility of achieving energy transfer from the excited semiconductor NPs. The competing excited state electron deactivation pathways are depicted in Scheme 1. The energy transfer process is dominant in pristine graphene sheets as shown by Brus and co-workers using confocal microscopy.⁴⁷ They observed quenching of band gap emission of CdSe by graphene via an energy transfer process with a rate constant of $9 \times 10^9 \text{ s}^{-1}$.

We have recently examined the excited state dynamics between CdSe quantum dots and GO/RGO with the aim of differentiating electron and energy transfer effects.⁴⁹ CdSe quantum dots anchored to GO and RGO exhibit strong fluorescence quenching. Due to the abundance of defect sites, both electron and energy transfer are expected to contribute to excited state deactivation in such systems. Once the CdSe–GO is subjected to visible illumination, we observe electron transfer resulting in the reduction of GO to RGO. Prolonged steady-state illumination of CdSe beyond the level required for reduction of GO causes electrons to accumulate within the RGO network. As charging ensues, further electron transfer is stunted, shutting off the electron-transfer-quenching pathway. Consequently, quenching in CdSe–RGO systems under prolonged illumination is dominated by energy transfer. The increased emission lifetime

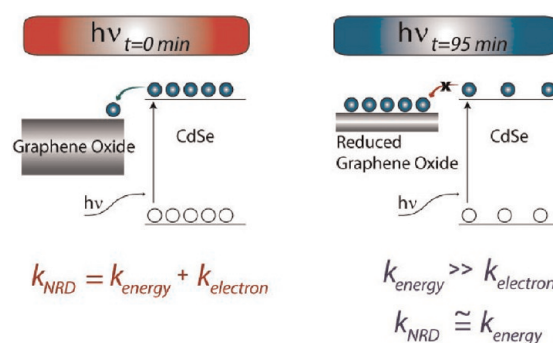


FIGURE 4. Electron transfer rates in CdSe quantum dot–RGO composites are manipulated by prolonged irradiation. (left) During initial illumination, photoexcited electrons in CdSe flow uninhibited to the GO. (right) Following prolonged illumination, transfer of photoexcited electrons from CdSe to RGO is stunted as electrons accumulate in RGO. Reprinted with permission from ref 49. Copyright 2012 American Chemical Society.

and fluorescence recovery of CdSe observed during steady-state illumination supports this argument (Figure 3).

Comparing the photoluminescence lifetimes of CdSe–GO and CdSe–RGO composites both before and after prolonged irradiation (Figure 4), one can obtain the rate constants for energy and electron transfer processes. The energy transfer and electron transfer rates obtained from CdSe to GO/RGO were estimated at $5.5 \times 10^8 \text{ s}^{-1}$ and $6.7 \times 10^8 \text{ s}^{-1}$, respectively.

Applications of Graphene-Based Semiconductor and Metal Nanoassemblies

Although the high density of defects in RGO makes it unattractive for purely electronic applications, they are considered to be useful in many other specialized applications due to the availability of high surface area and the ease

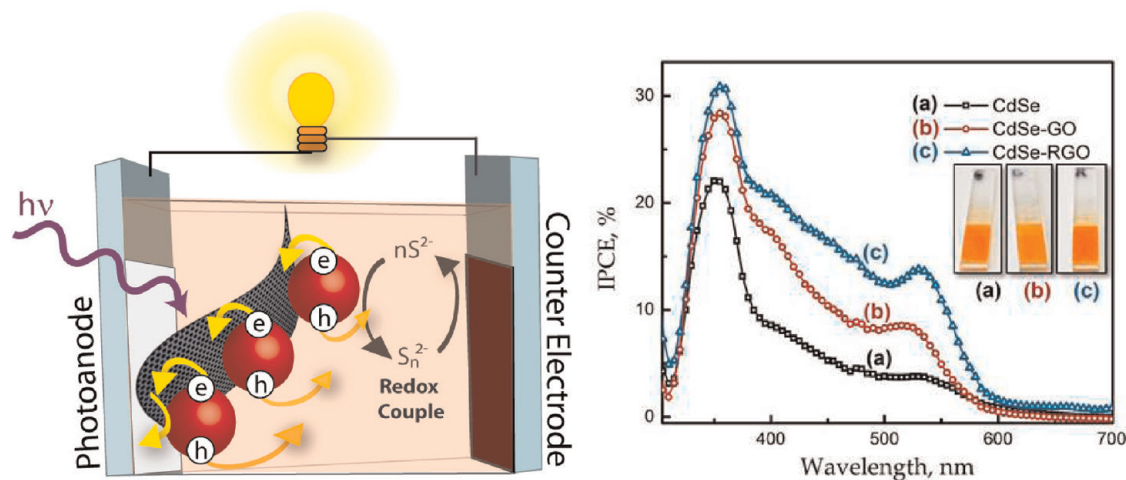


FIGURE 5. (left) Schematic depicting electron transfer from CdSe quantum dots to RGO where they are shuttled to the current collecting photoanode surface. (right) Incident photon to current efficiency (IPCE) spectra of CdSe-only, CdSe-GO, and CdSe-RGO photoanodes. (R)GO-CdSe composite electrodes show improved current collection at ~ 535 nm, the absorption peak for CdSe. This demonstrates the electron-mediating role of graphene in such mesostructured systems. Adapted and reprinted with permission from ref 49. Copyright 2012 American Chemical Society.

of bulk synthesis using chemical exfoliation. In particular, the availability of functional groups on the 2D carbon surface provides new opportunities to design organic-RGO or semiconductor- or metal-RGO composites. In energy-related applications such as solar cells and storage batteries, RGO serves as a platform to shuttle electrons efficiently, thus improving the overall performance of the devices.

Semiconductor quantum dot-RGO materials have also found applications in biomolecule sensing, bioimaging, LEDs, and flexible and transparent photoconductors.^{50–52} Initial work in this area has focused on the synthetic design of these composites. Approaches include chemical vapor deposition of quantum dots onto RGO, solution-based and electrochemical in situ growth of quantum dots on RGO, and ligand exchange in quantum dots to create more intimate connections with graphene.^{53–55}

Quantum Dot Solar Cells. The electron transfer properties described in the preceding sections suggest that semiconductor-RGO composite films designed for light energy conversion should reap the benefits of improved charge separation. For example, in the case of RGO incorporated into TiO₂ films, increased photoexcited electron transfer from TiO₂ NPs to the current collecting electrode is attributed to RGO's ability to assist in charge separation and shuttle electrons through its 2D conductive network.^{56,57}

Enhanced photoelectrochemical performance was also demonstrated using CdSe-RGO composites as photoanodes in quantum dot solar cells (QDSCs). Here RGO collects photogenerated electrons from excited CdSe quantum dots, suppressing recombination and facilitating electron

transport to the collecting electrode surface. The process is represented schematically in Figure 5, left panel. QDSCs constructed using CdSe-RGO photoanodes exhibit a marked improvement (up to $\sim 150\%$) in photocurrent response over CdSe-only photoanodes. The incident photon to current efficiency (IPCE) of the films confirms the increased photoresponse of the composite films and attributes the gain to an increased photocurrent contribution from the quantum dots (Figure 5, right panel). This further ascertains the beneficial role of RGO in improving QDSC performance.

Another key role for RGO in QDSCs lies in the development of new electrode materials, enabling metal-free realization of transparent conducting surfaces or serving as a network to relay redox processes tailored to specific applications.^{58–62} One such example involves the development of a Cu₂S-RGO composite counter electrode, which complements the common sulfide-polysulfide electrolyte redox couple used in QDSCs.⁶³ Poor performance by standard electrodes such as Pt in sulfide medium encumbers the performance of QDSCs, giving rise to poor fill factors of ~ 0.25 . Counter electrodes employing a Cu₂S-RGO composite material exhibit significant improvements in fill factors (>0.5). Benchmark efficiencies in the range of 4.4–5.4% were achieved using such counter electrodes.^{63,64}

Dye-Sensitized Solar Cells (DSSCs). Improved photoelectrochemical performance observed in DSSCs utilizing graphene-TiO₂ composites is attributed to higher dye loading, longer electron lifetimes, faster electron transfer, and lower recombination and back transfer observed in these

systems.^{65–67} Porphyrin-based photovoltaic architectures incorporating zinc porphyrin and ZnO NPs have been used as a model system to probe the effectiveness of RGO in promoting the charge separation and charge transport in dye-sensitized solar cells.²⁶ Specifically, RGO's ability to shuttle electrons can be used to promote charge transport within the mesoscopic TiO₂ network.^{56,66} The inorganic/organic architecture built with RGO/ZnO/porphyrin utilizes a unique electron transfer cascade beginning with photoexcitation of the zinc porphyrin. Photogenerated electrons in the zinc porphyrin are transferred to ZnO NPs then to RGO where they are shuttled to the SnO₂ layer on the electrode surface (Figure 6). Despite the multiple steps involved in the electron transfer process, these ternary systems exhibit efficient photocurrent generation, with incident photon to current efficiency approaching ~70%.

Energy Storage. Semiconductor–RGO architectures have also been employed as Li-ion battery anodes, capaci-

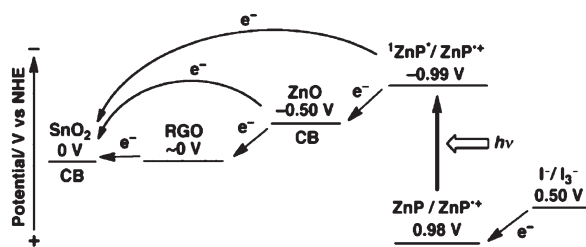


FIGURE 6. Band position diagram for the ternary electron transfer cascade achieved in ZnP–ZnO–RGO composites under illumination. Incident photon to current efficiency values of ternary composites yield efficiencies up to ~70%. Reprinted with permission from ref 26. Copyright 2011 American Chemical Society.

tors, and catalyst materials for hydrogen production.^{68–72} Lithium ion battery anode composites made with RGO–Mn₃O₄, –TiO₂, and –Co₃O₄ have recorded high specific capacities, faster charging rates, and good cycling stability.^{73,74} In the case of a MnO₂–RGO composite used as a Li-ion battery, improved kinetics at the electrode are attributed to faster Li⁺ diffusion rates and greater capacitance in the composite during discharge.⁷⁵

SERS Substrates. While previous findings suggest the benefit of using graphene as a surface-enhanced Raman scattering (SERS) substrate for trace molecule detection, the majority of reports in this area utilize the deposition of metal nanoparticles on graphene as a method to enhance graphene's Raman signals.^{76–78} A recent advancement in the ability to achieve side-selective deposition of semiconductor and metal NPs on RGO has resulted in RGO composite SERS sensors with photoactive control over metal nanoparticle composition, size, and loading (Figure 7, left panel).⁷⁹

These films consist of single-layer RGO sandwiched between a thin film of TiO₂ and an array of metal NPs. These semiconductor–graphene–metal (SGM) films utilize photocatalytic activity of sublayered TiO₂ NPs to induce reduction of Ag⁺ ions to Ag NPs on the side of RGO opposite the TiO₂. Under UV-illumination, photogenerated electrons in TiO₂ are transferred to and through the RGO sheet where they are used in the reduction of Ag⁺ ions. The electron transfer process demonstrates the ability of electrons to hop from one side of graphene to the other. In addition to segregating the semiconductor and metal NPs to opposite sides of the RGO, the photocatalytic process allows for control over the

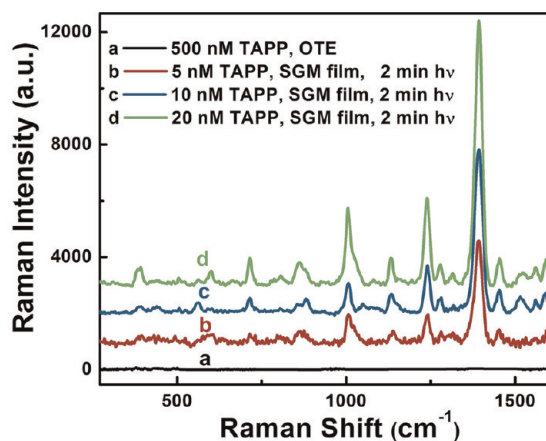
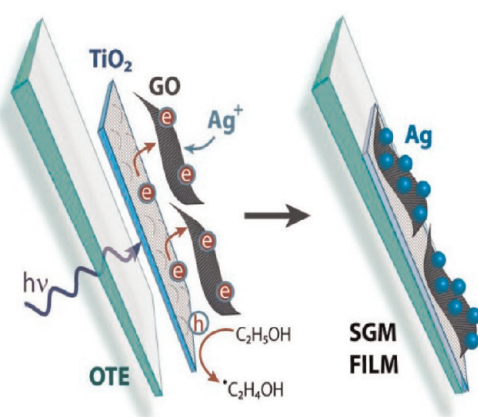


FIGURE 7. (left) Schematic illustration of the electron hopping process occurring in the production of semiconductor–graphene–metal (SGM) films. Under UV-illumination, electrons photogenerated in TiO₂ are transferred to the GO sheet. The GO sheet then utilizes the electrons to reduce Ag⁺ ions into Ag nanoparticles on the RGO side opposite the TiO₂. (right) SERS sensors made from SGM films achieve nanomolar detection levels of target molecule. Reprinted with permission from ref 79. Copyright 2012 American Chemical Society.

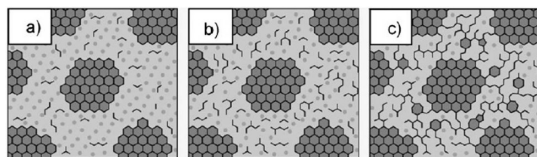


FIGURE 8. Series of schematics representing structural transitions from GO to RGO. (a) As-prepared GO sp^2 islands, functioning as luminescence centers, isolated by sp^3 regions, (b) slightly reduced GO showing initial restoration of sp^2 from sp^3 bonding, and (c) further reduction of sp^3 -bonded areas to a point where percolation between the sp^2 networks is restored. Reprinted with permission from ref 86. Copyright 2010 American Chemical Society.

size and loading of metal NPs while eliminating the need for extraneous chemical reductants.

In the presence of trace organic contaminants, the graphene in SGM films facilitates the accumulation of target molecules near the metal NP surface through adsorption. The interaction between organic molecules and graphene enables SGM films to attain a high degree of target-molecule concentration near the SERS-active metal crystallites, thus facilitating higher sensitivity. By this approach, 5 nM tetra-(4-aminophenyl) porphyrin (TAPP) was detected with a strong signal-to-noise ratio, which is illustrated in Figure 7, right panel. The SGM architecture represents a versatile template with the potential for assimilation into applications where the usefulness of the material depends on the ability to control and modify composition, size, and loading of nanoparticles.

Status and Future Outlook of RGO-Based Nanoarchitectures

It should be noted that in a majority of the reports examining RGO-based assemblies for light energy conversion, the role of RGO is more of a facilitator in the charge separation process and not as a direct absorber of light. Though not discussed in detail here, GO is often used as a hole transporting material in organic photovoltaics and light-emitting diodes.^{80,81} Although pristine graphene has a zero band gap, one can open a band gap and render p- or n-type semiconducting properties through functionalization.^{82–86} Even in these doped systems, the intrinsic band gap remains small and corresponds to transitions in the infrared. Hence their effectiveness to serve as an efficient light harvester remains negligible. Contrary to this argument, recent reports claim visible emission from chemically exfoliated GO, intimating higher energy transitions. It should be noted that smaller aromatic oligomers and PAHs (polyaromatic hydrocarbons), which exhibit molecular-like properties,

are likely the contributors to this visible (high energy) emission.⁸⁷ The excitation wavelength-dependent emission observed in these studies indicates the presence of many different molecular-like species (Figure 8).⁸⁶ Caution should be exercised when claiming photocatalytic activity of GO with direct excitation. Careful evaluation of impurities or other coexisting species present in the system should be analyzed before drawing such conclusions.

The representative examples discussed in this Account have just begun to tap into the potential of RGO as a large surface area catalytic platform to manipulate photoactive composite assemblies for light energy conversion, energy storage, and other useful applications. Its favorable energetics for intercepting excited states and inducing charge separation, along with the ease of developing electrode materials with tailored properties make RGO an exceptionally important mediator for improving the performance of solar cells and photocatalyst arrays.

By extension, one can imagine the possibility of utilizing semiconductor–graphene–metal NP composites as stand-alone, transferable films to accomplish selected photochemical catalytic reactions. It also is conceivable to engineer RGO composite films that separate different catalyst NPs and their respective functions on opposing sheet surfaces. This configuration could be exploited to separate half-reactions or enable a multioperational response from RGO-supported catalyst arrays.

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BIOGRAPHICAL INFORMATION

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FOOTNOTES

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