

## Collaborative research: Nano-manufacturing reduced graphene oxide

NSF Grant CMMI-1100290

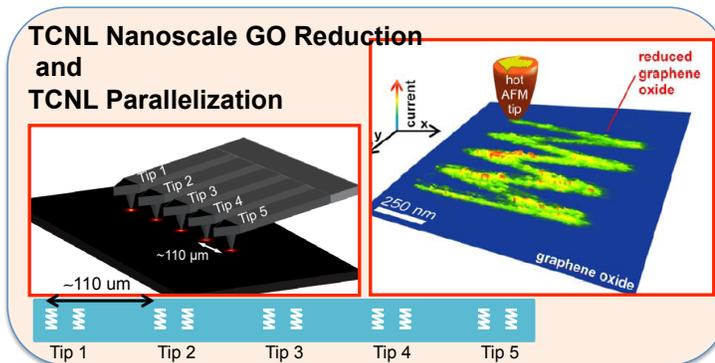
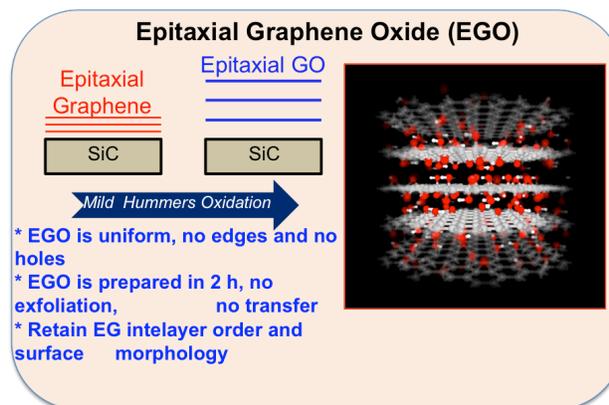
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Graphene and graphene-based materials hold great promise for the next generation of nanodevices. One of the most pressing issues for the technological use of graphene is the possibility to control physical and chemical properties by means of ad hoc functionalization. Thermal, chemical and optical reduction of graphene oxide have been explored as a route to produce graphene-based materials with the desired electron transport, mechanical and optical properties. Graphene oxide is indeed a material of great interest for its potential applications in nanoelectronics, nanoelectromechanical system, sensors, polymer composites, catalysis, energy storage devices and optics. However, the chemistry of graphene oxide and its response to external stimuli such as temperature and light are not well understood and only approximately controlled. This understanding is crucial to enable future applications of this material.

In this research project by combining experiments and density functional theory investigations we have discovered a **new type of graphene oxide films** [1-4]. **Ultra-thin epitaxial graphene oxide (EGO) films with a controlled number of layers were obtained by a mild Hummers oxidation of epitaxial graphene grown on silicon carbide substrates.** EGO films possess the following attributes. (1) EGO synthesis is rapid (~2 hours), the chemical oxidation process does not change the number or disrupt the order of the graphene layers, and the oxide films exhibit reproducible chemical and structural features. (2) Pristine graphene and EGO films exhibit comparable vertical inter-layer structural quality (coherence lengths and interlayer registry). (3) EGO films exhibit large interlayer separations (~9-10 Å) and – unexpectedly – minimal concentrations of water molecules intercalated in the multilayer structure. (4) EGO films are extremely smooth over the entire SiC substrate area, do not present holes or edges and they are uniform, showing no propensity to exfoliate. (4) The EGO film structure – chemical features and interlayer spacing – is stable up to temperatures of 140°C and in N<sub>2</sub> atmosphere.

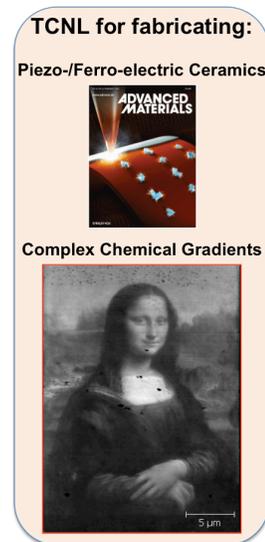
Overall, our work provides experimental information on an interesting and potentially transformative graphene oxide material, as well as fundamental understanding on the relationship between interlayer spacing, water content, and intralayer structure in this



or other forms of graphene oxide.

Furthermore, in this project we have explored the potential of a new lithography technique invented at Georgia Tech by the PIs of this proposal, thermochemical nanolithography [5]. **Thermochemical nanolithography (TCNL)** employs heated nano-size tips to induce local thermally activated chemical reactions to change the chemical functionality of surfaces, or obtain new nanostructures. Examples of thermally activated reactions possible with TCNL are deprotection of functional groups at surfaces, reduction of oxides and crystallization of amorphous materials. TCNL is based on a scanning probe microscopy (SPM) approach, where nano-size tips can be moved in three dimensions with a spatial resolution as low as 1 nm. This technique offers advantages in terms of the combination of high speed, high resolution, material flexibility, potential towards parallelization, and the versatility of working under ambient conditions.

During the development of this NSF project we have demonstrated the ability to reduce graphene oxide at the nanoscale by using TCNL [6]. The resulting nanostructures have a conductivity that can be tuned over 4 orders of magnitude. Furthermore, we have demonstrated the ability of TCNL to create chemical gradients with sub-100 nm resolution [7], and to fabricate complex nanostructures of ferroelectric/piezoelectric PZT and PTO ceramics [8]. More recently, we have worked on the parallelization of TCNL by fabricating and using arrays with 5 tips [9].



## References

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