Three Dimensional Graphene-based Assemblies

Marcus A. Worsley, Matthew D. Merrill, Elizabeth Montalvo, Patrick Campbell, Jianchao Ye, Supakit Charnvanichborikarn, Swanee Shin, Monika Biener, Sergei O. Kucheyev, Michael Stadermann, Yinmin Wang, Theodore F. Baumann, Juergen Biener

> Physical Sciences Directorate, Lawrence Livermore National Laboratory 7000 East Ave, Livermore, CA, 94551 United States

Graphene has shown the potential to significantly impact a number of different technologies, including energy storage. Properties such as high surface areas and electrical conductivity make it a promising material for hydrogen storage, battery, and ultra capacitor applications. One route to realizing the full potential of graphene in energy storage applications is the assembly of three-dimensional macroscopic graphene networks that retain the properties of individual graphene sheets. Herein we report the assembly of graphene sheets into a hierarchical architecture with length scales extending from the nanoscale to the macroscopic regime. These graphene macroassemblies are formed via cross-linking reactions between single- and/or few-layer graphene oxide (GO) sheets in suspension. The hierarchical structure possesses a number of novel properties including mechanical stiffness (up to 10 GPa) and electrical conductivities (up to 10^5 S/m) orders of magnitude higher than previously reported, surface areas that approach the theoretical values expected for a single graphene sheet (~2500 m²/g), and extraordinarily large mesopore volumes (up to 5 cm³/g). Energy storage behavior for capacitor and Li-ion battery applications were evaluated. The graphene-based electrode simultaneously exhibited high energy (~ 10^2 Wh/kg) and power densities (~ 10^2 kW/kg) in aqueous electrolytes (symmetric cell), while metal oxide-coated graphene cathodes exhibited large Li-ion capacities (~1000mAh/g). The details of the synthesis and characterization of these novel graphene assemblies will be presented.