Preparation of ultra-light, "shell-less" graphene aerogel via simple pre-reduction method

Kaiwen Hu¹, Xingyi Xie^{1,2}, Marta Cerruti¹ and Thomas Szkopek³

¹Department of Mining and Materials Engineering, McGill University, Montreal, QC, H3A 2B2, Canada. ²College of Polymer Science and Engineering, State Key Laboratory of Polymer Materials and Engineering, Sighuan University, Changdy, Sighuan, 610065, Ching

Engineering, Sichuan University, Chengdu, Sichuan, 610065, China.

³Department of Electrical and Computer Engineering, McGill University, Montreal, QC, H3A 0E9,

Canada.

Kaiwen.hu@mail.mcgill.ca

Reduced graphene oxide (RGO) gels are emerging three dimensional graphene macroscopic assemblies of potential use in many applications including energy storage, pollutant adsorption and gas sensing ¹. Conventional RGO gels are often prepared via solution based gelation involving the reduction of graphene oxide (GO) through chemical or solvothermal reduction. Often, the gel is formed with a shell structure enveloping the interior porous bulk ². The conventional gel also shows high volume shrinkage and is thus mechanically rigid, demonstrating severe plastic deformation upon compression. This makes them less attractive to applications such as reusable oil sponges, energy absorbing materials and electromechanical sensors.

In this work, we present a simple pre-reduction technique prior to hydrothermal gelation of GO that allows us to prepare RGO aerogels with ultra-low density and excellent mechanical elasticity. The prereduction step also weakens the surface anchoring of GO at interfaces resulting an open structure with the porous bulk structure immediately accessible from the surface ². These new features make the prereduced RGO aerogel more attractive to the aforementioned applications.

References

1. Cong, H.-P.; Chen, J.-F.; Yu, S.-H., Graphene-based macroscopic assemblies and architectures: an emerging material system. *Chemical Society Reviews* **2014**, *43* (21), 7295-7325.

2. Hu, K.; Xie, X.; Cerruti, M.; Szkopek, T., Controlling the Shell Formation in Hydrothermally Reduced Graphene Hydrogel. *Langmuir* **2015**, *31* (20), 5545-5549.

Scheme 1. Graphene hydrogel (GHG) formation under hydrothermal conditions: a) conventional GHG with shell; b) GHG by thermal or chemical pre-reduction prior to hydrothermal treatment.



Figure 1. Optical and SEM images of conventional and shell-less GHG. a) Photograph of conventional GHG, b) shell-less GHG prepared via thermal pre-reduction and c) shell-less GHG prepared via chemical pre-reduction using vitamin C. SEM images of the exterior (d) and interior (e) structure of shell-less GHG in b) with similar microstructure.