Supplementary Information

Evolution of Heterogeneity and Chemical Functionality during the Oxidation of Graphite

Additional Details of Oxidation Procedure

A 4 liter beaker (graduated, PYREX® Griffin) was used for the graphene oxide synthesis. Dissolution of sodium nitrate took 30 minutes and raised the temperature of the solution to 40 °C. Temperature then slowly rose to 80 °C during the addition of the oxidant. After complete addition of the oxidant, the color of the solution turned to dark green.

For quenching the GO synthesis reaction, we used the common approach of adding DI water and hydrogen peroxide (Acros Organics, 35 wt. %), and the solution turned yellow. The quenching procedure was kept consistent for all the GO samples.

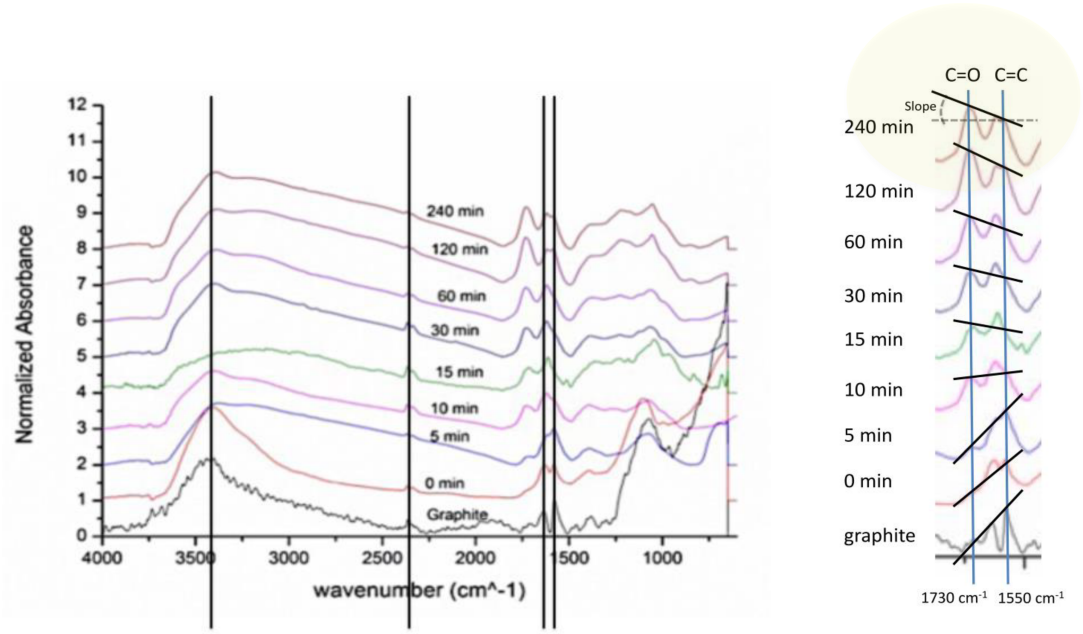

Figure S1. Left picture: FT-IR spectra of GO samples sampled at different times. The vertical lines on the right are at the wave numbers used for determining the peak heights used for the black curve in Figure 2B in the manuscript. The two vertical lines on the left indicate that the various spectra are in alignment; something can be difficult to see with the unaided eye. Right picture: Formation of the line (in black) joining the two peak maxima (C=O & C=C in this case) for all FTIR spectra, and calculation of slope of that line (demonstrated for 240 min FTIR spectrum).

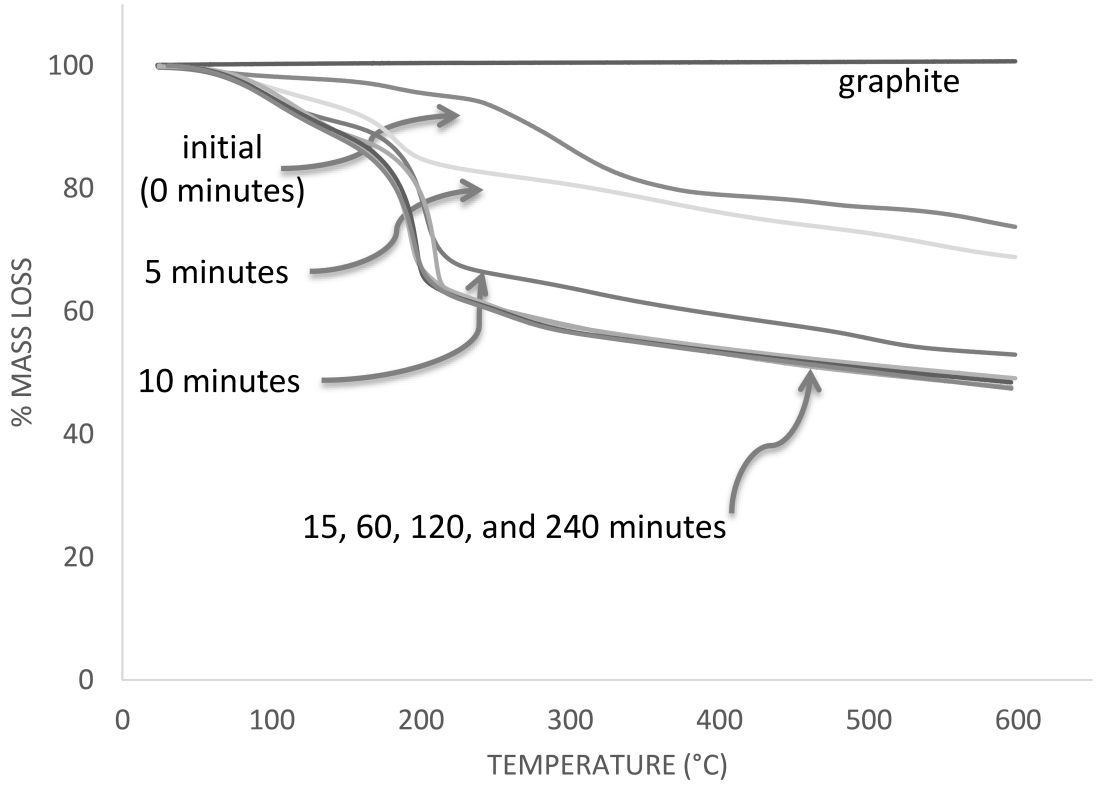

Figure S2. TGA showing increased mass loss with increasing oxidation time

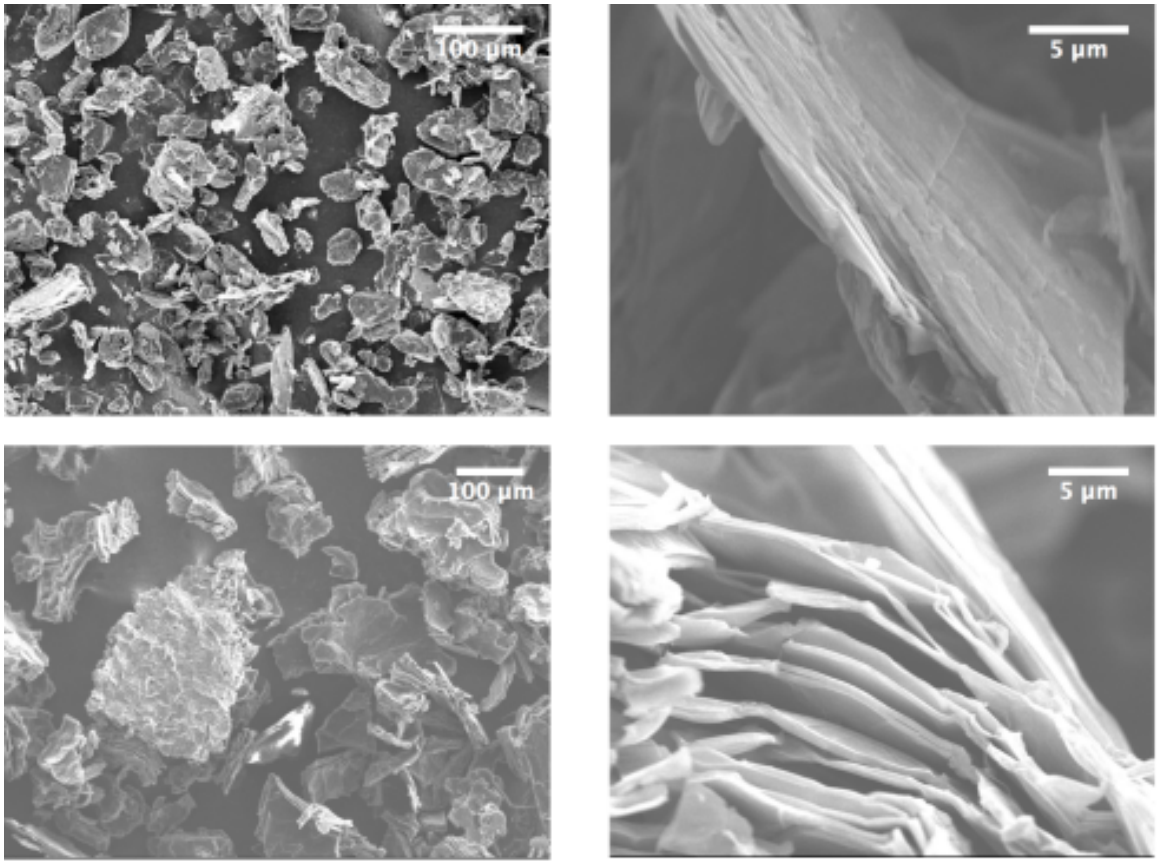

Figure S3. SEM images of short oxidation time samples, (a,b) unreacted graphite, and (c,d) 0-minute oxidation.

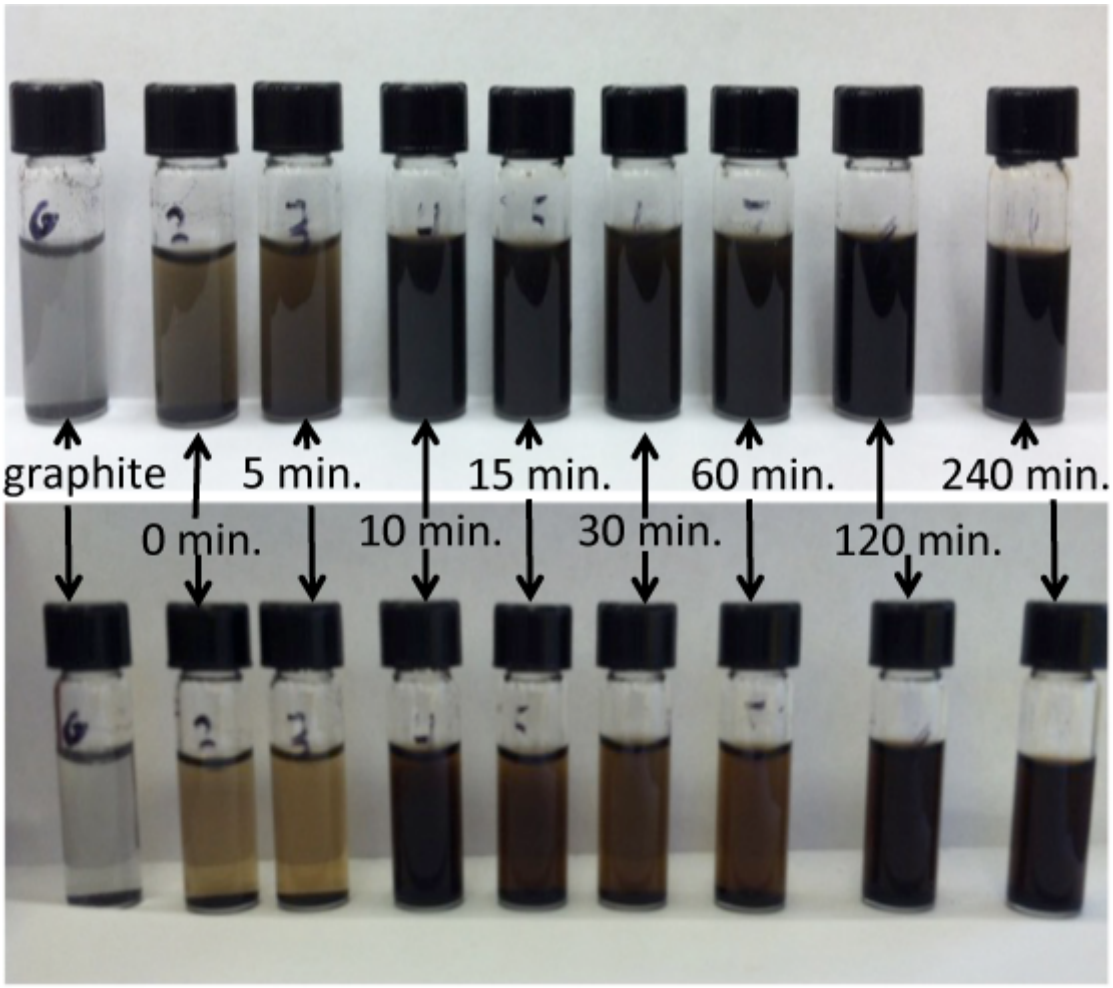

Figure S4. Dispersions of GO in water from graphite (left) with increasing oxidation time up to 4 hours (right) immediately after sonication (top) and after 5 days of settling (bottom).