

Electronic Supplementary Information (ESI)

Molecular force transfer mechanisms in graphene oxide paper evaluated using atomic force microscopy and *in situ* synchrotron micro FT-IR spectroscopy

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X-ray diffraction of GO paper

The evaluation of exfoliated graphene oxide and the inter-layer spacing of GO paper are evident from its X-ray diffraction pattern (XRD), as comparison, the raw graphite powder sample was also measured.

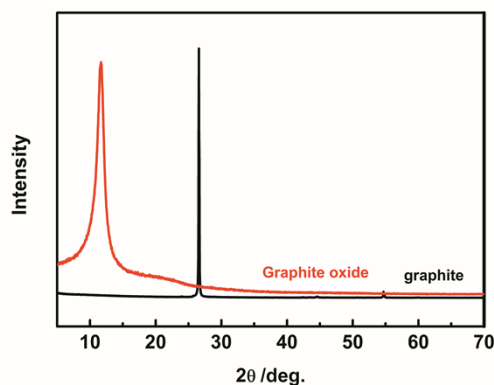


Fig. S1 XRD of the raw graphite powder and produced GO paper

The diffraction peak at $2\theta = 26.5^\circ$ is shown for the graphite powder in Fig. S1 and is typical for graphite material, which corresponds to the inter-layer spacing of 0.335 nm for graphite layers. The GO paper exhibits a diffraction peak at $2\theta = 11.6^\circ$, which corresponds to an inter-layer spacing of approximately 0.8 nm and is the expected XRD feature for exfoliated GO sheets [S1]. The absence of a strong graphitic XRD peak in the GO paper suggests a lack of graphitic clusters.

AFM images of fabricated GO sheets

The semi-contact AFM (NT-MDT NTegra, Rus.) images are shown below to demonstrate the distribution and thickness of the fabricated GO sheets.

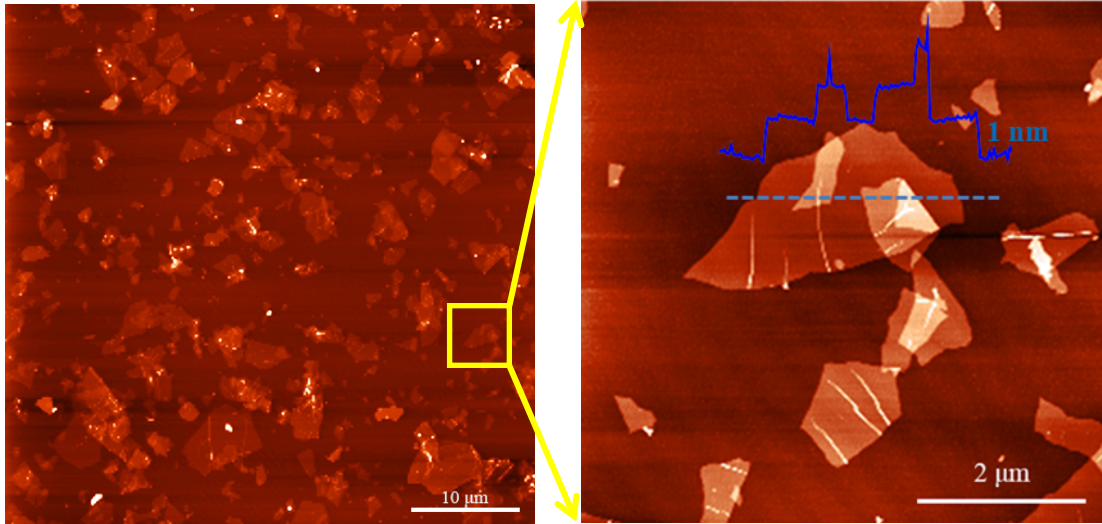


Fig. S2 Semi-contact mode AFM images of GO sheets on an atomically flat mica substrate in different magnifications, with an individual GO sheet height profile along the blue line shown on the right image.

More than 90 % of the fabricated GO sheets in Fig.S2 were exfoliated completely with a measured thickness around 1 nm and average size around 1-2 μm , indicating the starting GO paper predominantly consisted of single layer graphene oxide sheets rather than unexfoliated multilayer graphite oxide flakes.'

Proposed model to describe coupling between C=C and C=O bonds in GO paper

According to Hooke's law, the force is constant for two springs connected in series, and the bond strain varies inversely with the force constant according to:

$$F = -k_{C=C}\Delta\varepsilon_{C=C} = -k_{C=O}\Delta\varepsilon_{C=O} \quad (\text{S1})$$

where $k_{C=C}$ and $k_{C=O}$ are the force constants for C=C and C=O bond types and $\Delta\varepsilon_{C=C}$ and $\Delta\varepsilon_{C=O}$ are the corresponding bond strains. The relationship between the bond strain rate with

applied strain can be stated as a function of the ratio between the force constants of the C=C and C=O bonds using Equation (S1) to give:

$$\frac{d\varepsilon_{C=O}}{d\varepsilon_{applied}} = \frac{k_{C=C}}{k_{C=O}} \frac{d\varepsilon_{C=C}}{d\varepsilon_{applied}} \quad (S2)$$

Further modification of the above equation to incorporate the effect of the orientation θ of the C=O bond relative to the C=C bond and their resultant force constant ratio gives:

$$\frac{d\varepsilon_{C=O}}{d\varepsilon_{applied}} \cos \theta = \frac{k_{C=C}}{k_{C=O}} \frac{d\varepsilon_{C=C}}{d\varepsilon_{applied}} \quad (S3)$$

Therefore, a function that describes the C=O bond strain with applied strain can be modeled with the relative angle between C=C and C=O bond as the variables,

$$\frac{d\varepsilon_{C=O}}{d\varepsilon_{applied}} = f(\cos \theta) = \frac{k_{C=C}}{k_{C=O}} \frac{d\varepsilon_{C=C}}{d\varepsilon_{applied}} \frac{1}{\cos \theta} \quad (S4)$$

C=O bond peak full width at half maximum (FWHM) and peak intensity during the mechanical testing

The C=O bond orientation angle represents all of the bonds within the volume of the sample probed by the IR and is due to either an average angle over a wider distribution of C=O bonds or the common C=O bond angle in the GO material. The distribution of C=O bonds can be tested by recording the bond angle distributions using bond peak full width at half maximum (FWHM) broadening with peak intensity from FTIR. A large distribution in the C=O bond angles would be expected to cause peak broadening, and a resultant increase in the FWHM, due to bonds aligned with the applied external force strain more than mis-aligned bonds with relatively large θ values. Fig. S3a shows the C=O peak FWHM with applied strain and highlights little variation in the FWHM during mechanical deformation of the GO

sample, suggesting a relatively small distribution of C=O bond angles. The C=O peak intensity with applied strain is additionally shown in Fig. S3b and exhibits little peak intensity variation. A constant C=O peak intensity indicates the number of bonds evaluated within the plane of the FTIR polarization during strain remains constant during mechanical testing. This data indicates a static C=O structure during mechanical deformation of the GO paper and suggests that the weak inter-sheet bonding is insufficient to cause bond orientation along the direction of the applied load.

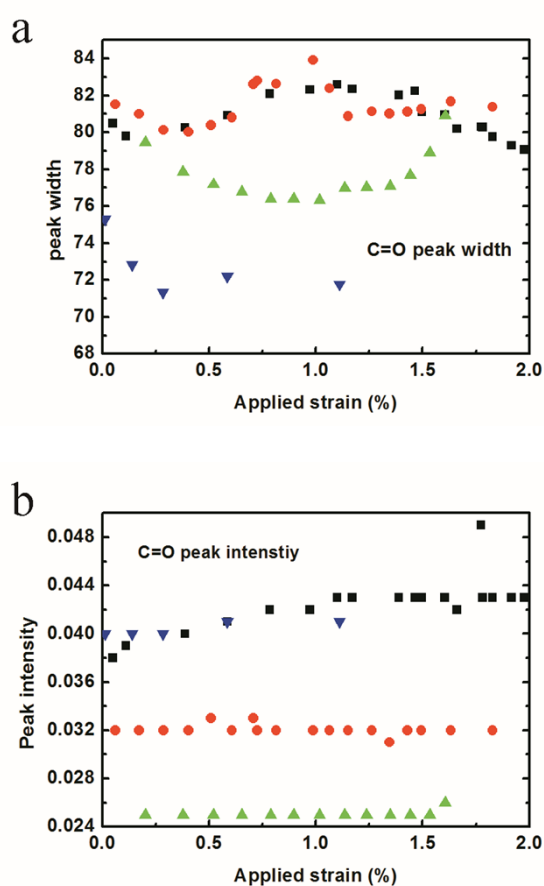


Fig. S3 (a) C=O bond peak width with applied strain. (b) C=O bond peak intensity with applied strain.

Reference

[S1] D. A. Dikin, S. Stankovich, E. J. Zimney, R. D. Piner, G. H. B. Dommett, G. Evmenenko, S. T. Nguyen and R. S. Ruoff, *Nature*, 2007, **448**, 457.