

# Structure Evolution and Control in Graphene Polymer Composites

**Karl S. Coleman<sup>1</sup>**

Stephen C. Boothroyd<sup>1</sup>, David W. Johnson<sup>1</sup>, Michael P. Weir<sup>2</sup>, Carl D. Reynolds<sup>2</sup>, James M. Hart<sup>1</sup>, Andrew J. Smith<sup>3</sup>, Nigel Clarke<sup>2</sup>, Richard L. Thompson<sup>1</sup>

<sup>1</sup>Department of Chemistry, Durham University, Durham, DH1 3LE, UK. <sup>2</sup>Department of Physics and Astronomy, University of Sheffield, Sheffield, S3 7RH, UK. <sup>3</sup>Diamond Light Source Ltd., Diamond House, Harwell Science and Innovation Campus, Didcot, UK.

[k.s.coleman@durham.ac.uk](mailto:k.s.coleman@durham.ac.uk)

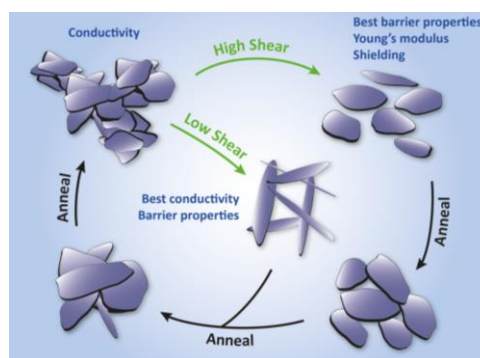
Graphene and related 2D materials have extraordinary physical properties that along with their high aspect ratio make them excellent candidate filler materials for polymer nanocomposites, capable of producing significant gains in material properties at extremely low concentrations. We have recently investigated the conformations of polymer chains in polymer-graphene nanocomposites, and have shown that the chains have a reduced radius of gyration, and reduction in interchain entanglements, that is consistent with confinement at a solid interface.[1] We have also discovered that when we look at the graphene filler we find, using scattering techniques, that the host polymer matrix can influence the morphology of the graphene.[2] With both of these in mind we have turned our attention to the processing of graphene polymer composites. Reliable processing of such materials with uniform and consistent properties remains a significant challenge because of the difficulty in controlling the graphene conformation and dispersion. Using shear rate and shear history we can control graphene network morphology and nanocomposite properties. Remarkable changes in electrical impedance unique to composites of graphene nanoplates (GNPs) are observed. Low shear rates  $\leq 0.1 \text{ s}^{-1}$  break up the typical GNP agglomerates found in graphene composites, partially exfoliate the

GNPs to fewer graphene layers and reduce orientation, enhancing electrical conductivity in the composite materials. Whereas, at higher shear rates GNP orientation increases and the conductivity reduces by four orders of magnitude, as the graphene filler network is broken down. Interestingly the structure of the composite continues to evolve over time, reflected in further changes in conductivity, after the shear force has been removed and the process temperature maintained, figure 1. This work provides critical insights for understanding and controlling GNP orientation and dispersion within composites and will have important consequences in the industrial processing of graphene polymer composites via the informed design and choice of processing.

## References

- [1] Authors, M. P. Weir, D. W. Johnson, S. C. Boothroyd, R. C. Savage, R. L. Thompson, S. M. King, S. E. Rogers, K. S. Coleman, N. Clarke, *ACS Macro Lett.*, 5 (2016), 430–434.
- [2] M. P. Weir, D. W. Johnson, S. C. Boothroyd, R. C. Savage, R. L. Thompson, S. R. Parnell, A. J. Parnell, S. M. King, S. E. Rogers, K. S. Coleman, N. Clarke, *Chem. Mater.*, 28 (2016), 698–1704.

## Figures



---

**Figure 1:** Schematic of processing effects on the GNP structure within the composite

---