Size Effects during Transfer of Stress from Polymer to Graphene in Nanocomposites

G. Anagnostopoulos¹, Ch. Androulidakis¹, Z. Li², I. A. Kinloch², R. J. Young², K. S. Novoselov³, C.Y. Lu⁴, J. Parthenios¹, K. Papagelis^{1,5,*} and C. Galiotis^{1,6,*}

¹FORTH/ICE-HT, P.O. Box 1414, Patras GR-26504, Greece

²School of Materials, Univ. of Manchester, Grosvenor Street, Manchester, M13 9PL, UK

³ School of Physics and Astronomy, Univ. of Manchester, Oxford Road, Manchester, M13 9PL, UK

⁴ BGT Materials Limited, 2.312 Photon Science Institute, University of Manchester, Oxford Road, Manchester, M13 9PL, UK

⁵ Department of Materials Science, University of Patras, Patras GR-26504, Greece

⁶ Department of Chemical Engineering, University of Patras, Patras GR-26504, Greece

*Corresponding authors:

K. Papagelis, Department of Materials Science, University of Patras, Patras GR-26504, Greece (kpapag@upatras.gr)

C. Galiotis, Department of Chemical Engineering, University of Patras, Patras GR-26504, Greece, (c.galiotis@iceht.forth.gr)

Higher mechanical properties of graphene sheets have attracted increasing attention worldwide. Similar to other composites, the mechanical properties depend on the concentration, aspect ratio and distribution of the nanofiller in the matrix and the interface bonding¹. Besides simple reinforcing effects (Young's modulus and fracture strength), improvements in fracture toughness, fatigue strength and buckling resistance have also been reported in graphene-polymer nanocomposites²⁻³.

However, the underlying strengthening and toughening mechanisms are still not well understood. Several factors, such as interfacial adhesion, spatial distribution and alignment of graphene nano-filler are considered to be crucial for effective reinforcement in the nanocomposites. On the other hand, the graphene sheets such as CVD monolayer graphene films simply-supported or embedded in polymer matrix may create wrinkled structures that tend to unfold rather than stretch under applied loading⁴⁻⁶.

In this work, certain examples of size effects on the stress transfer characteristics in graphene/ polymer nanocomposites will be presented. The first one of those refers to stress transfer in exfoliated monolayer graphene in which it will be shown that the unintentional chemical doping of the graphene edges can affect the size of the stress transfer zone. Indeed for efficient stress transfer, flakes much greater than ~ 10 μ m are required. The second example refers to commercial CVD graphene either simply-supported and/or embedded in different polymer substrates such as poy(methyl methacrylate) (PMMA) and poly(ethylene terephthalate)(PET). For this case, the extractedstrain sensitivity is considerably lower than that of mechanically exfoliated graphene samples embedded into polymers. The unusualdeformation behaviour is attributed to the microstructure of the CVD graphene comprising ofmechanically isolated graphene islands separated by wrinkle material which, in turn, affectsconsiderably the stress transfer efficiency from the matrix to CVD graphene membrane.

References

1. Vallés, C.; Abdelkader, A. M.; Young, R. J.; Kinloch, I. A., The effect of flake diameter on the reinforcement of few-layer graphene–PMMA composites. *Composites Science and Technology* **2015**,*111* (0), 17-22.

2. Rafiee, M. A.; Rafiee, J.; Srivastava, I.; Wang, Z.; Song, H.; Yu, Z.-Z.; Koratkar, N., Fracture and Fatigue in Graphene Nanocomposites. *Small* **2010**,*6* (2), 179-183.

3. Rafiee, M. A.; Rafiee, J.; Wang, Z.; Song, H.; Yu, Z.-Z.; Koratkar, N., Enhanced Mechanical Properties of Nanocomposites at Low Graphene Content. *Acs Nano* **2009**,*3* (12), 3884-3890.

4. Xu, C.; Xue, T.; Guo, J.; Qin, Q.; Wu, S.; Song, H.; Xie, H., An experimental investigation on the mechanical properties of the interface between large-sized graphene and a flexible substrate. *J Appl Phys* **2015**,*117* (16), 164301.

5. Shen, X.; Lin, X.; Yousefi, N.; Jia, J.; Kim, J.-K., Wrinkling in graphene sheets and graphene oxide papers. *Carbon* **2014**,*66* (0), 84-92.

6. Li, Z.; Kinloch, I. A.; Young, R. J.; Novoselov, K. S.; Anagnostopoulos, G.; Parthenios, J.; Galiotis, C.; Papagelis, K.; Lu, C.-Y.; Britnell, L., Deformation of Wrinkled Graphene. *Acs Nano* **2015**.

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