**Supporting Information**

Effect of Folded and Crumpled Morphologies of Graphene Oxide Platelets on the Mechanical Performances of Polymer Nanocomposites

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The Halpin-Tsai model can be successfully applied to predict the modulus of GO based nanocomposites. For randomly oriented GO sheets in a polymer matrix, the composite modulus *EC* is given by

$E\_{c}=[\frac{3}{8}\frac{(2L\_{G}/3T\_{G})η\_{L}V\_{G}}{1-η\_{L}V\_{G}}+\frac{5}{8}\frac{1+2η\_{L}V\_{G}}{1-η\_{L}V\_{G}}]E\_{P}$ (1)

$η\_{L}=\frac{(E\_{G}/E\_{P})-1}{E\_{G}/E\_{P}+2L\_{G}/3T\_{G}}$(2)

$η\_{T}=\frac{(E\_{G}/E\_{P})-1}{E\_{G}/E\_{P}+2}$ (3)

where, *EC*, *EG*, and *Ep* represent the tensile modulus of the composites, chemically reduced GO sheet (0.25 TPa), [Ref1] and the polymer (2.72 GPa) respectively. *LG* is the lateral size of the GO sheets, *TG* is their thickness, and *VG* is the volume fraction of GO in the composite.



*Figure S1.* Young’s moduli of nanocomposites as a function of aspect ratio of GO sheets based on the Halpin–Tsai theoretical model.

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*Figure S2.* Images of 0.3 vol.%*l*-GO-based GO/PVA nanocomposites characterized by confocal laser scanning microscopy.



*Figure S3.* Shift of G-(a,b) and G+(c,d) peak positions as a function of applied strain for *l*- and *s*- GO/PVA nanocomposites under loading and unloading conditions.

**REFERENCES**

1. Gómez-Navarro C.; Burghard M.; Kern K. Elastic Properties of Chemically Derived Single Graphene Sheets. Nano Lett. 2008, 8, 2045-2049.