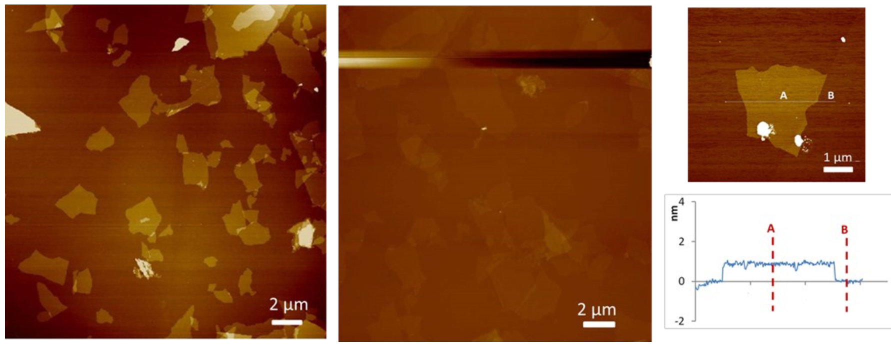
Electronic Supplementary Material (ESI) for RSC Advances.

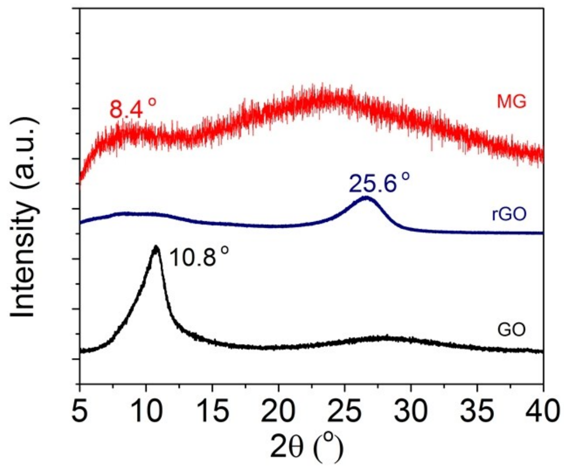
Spray-Coated Epoxy Barrier Films Containing High Aspect Ratio Functionalized Graphene Nanosheets

Peng Li[1], Tsao-Cheng Huang[1], Kevin L. White[1], Spencer Hawkins[1], Masaya Kotaki[2], Riichi Nishimura[3], and Hung-Jue Sue[1]*

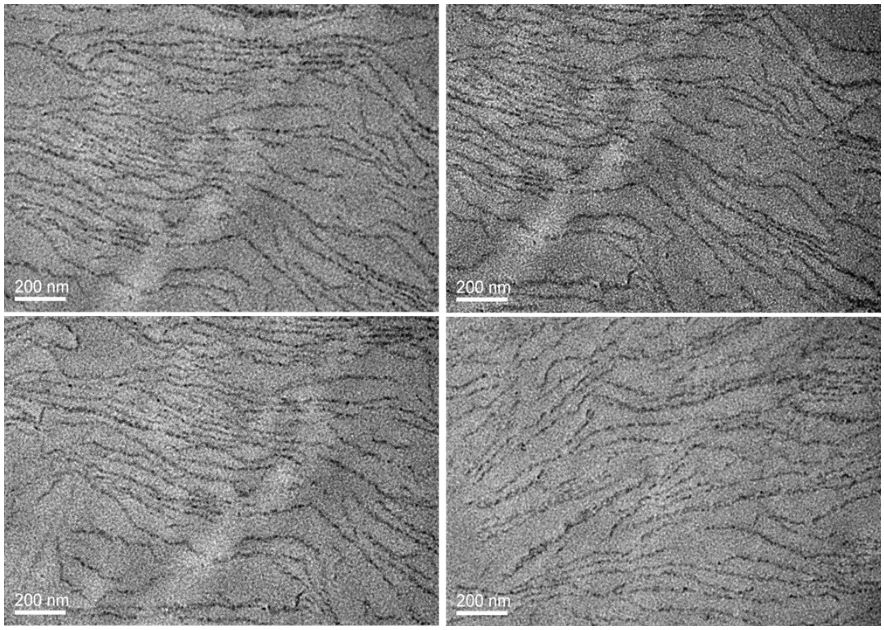
[1] Department of Materials Science and Engineering, Texas A&M University, College Station, TX 77843, United States.
[2] Material Research Center, Kaneka Americas Holding, Inc., College Station, TX 77843
[3] Frontier Materials Development Laboratories, Kaneka Corporation, Osaka 530-8288, Japan
*E-mail: hjsue@tamu.edu (H.-J. Sue).



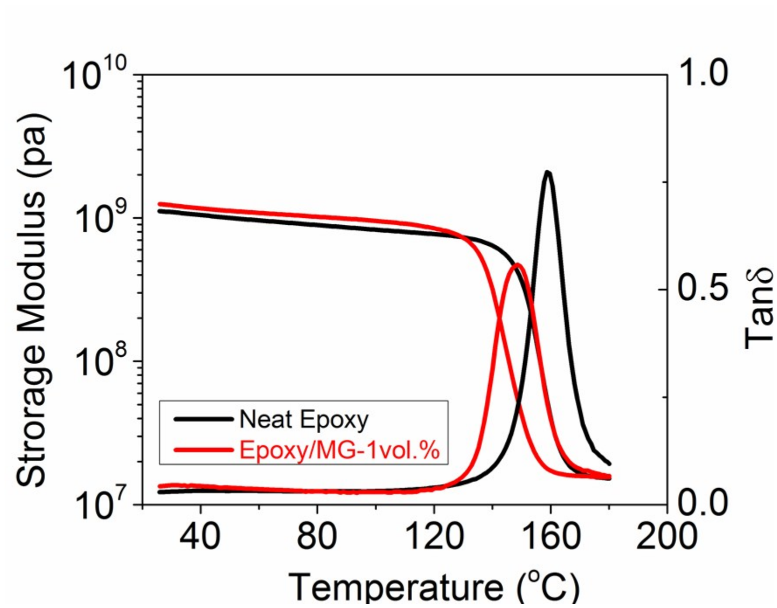
Supplemental Figure S1. Tapping-mode AFM image and height profile of MG.



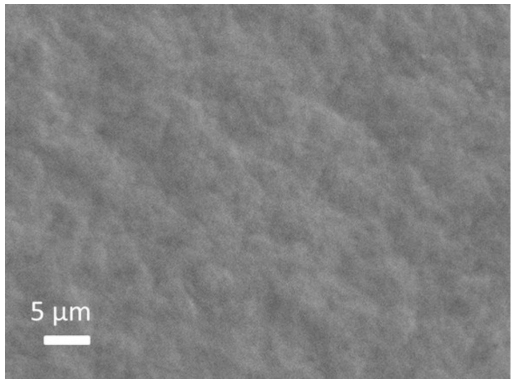
Supplemental Figure S2. XRD of graphene oxide (GO), reduced graphene oxide (rGO) and modified graphene (MG).



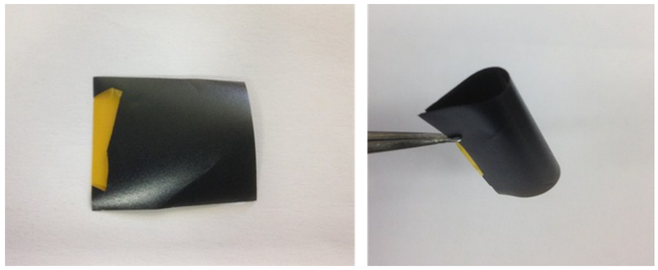
Supplemental Figure S3.TEM of cross-sectional epoxy/MG films (3.6 vol%) at different regions.



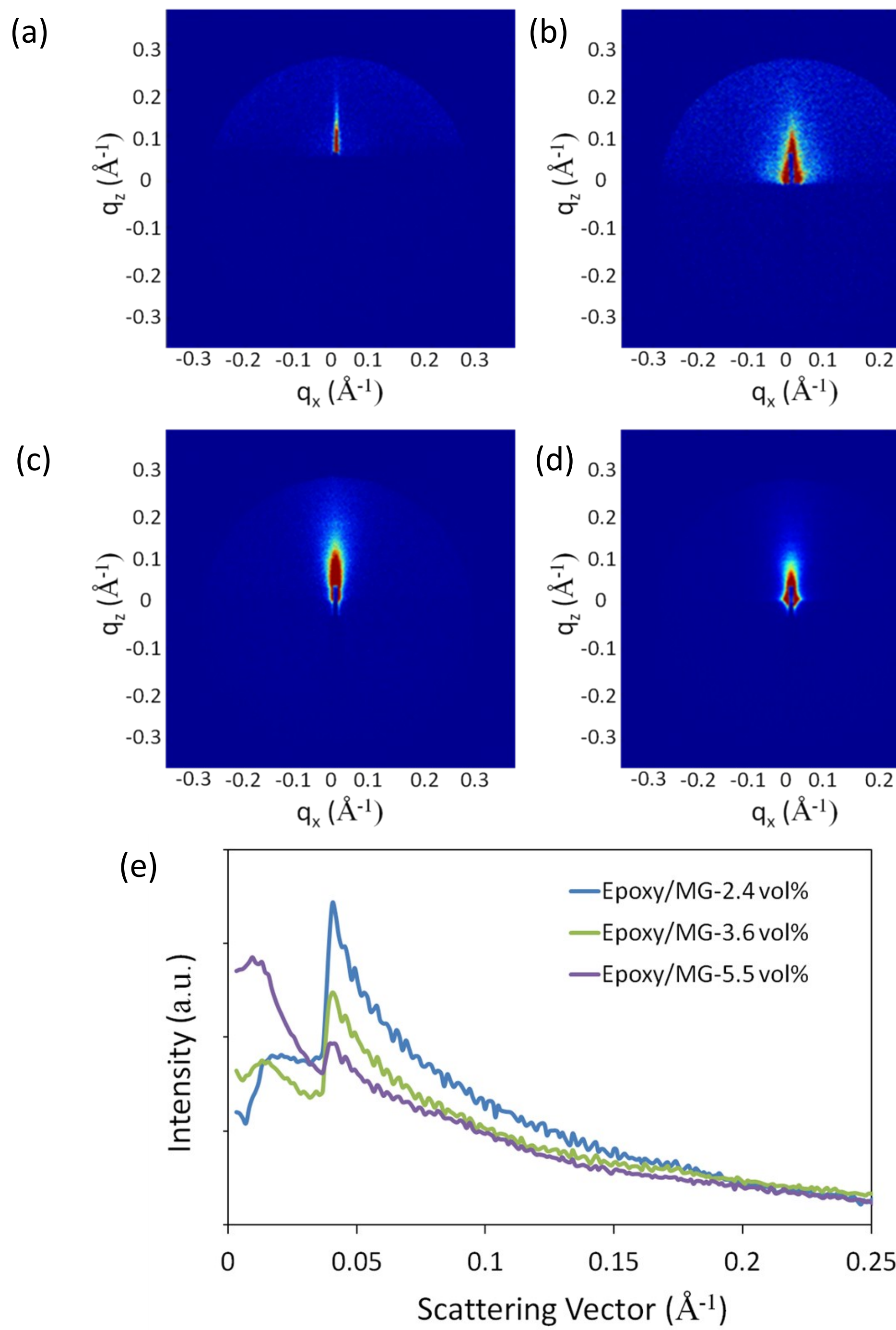
Supplemental Figure S4. DMA of neat epoxy and epoxy/MG (1vol.%) bulk samples.



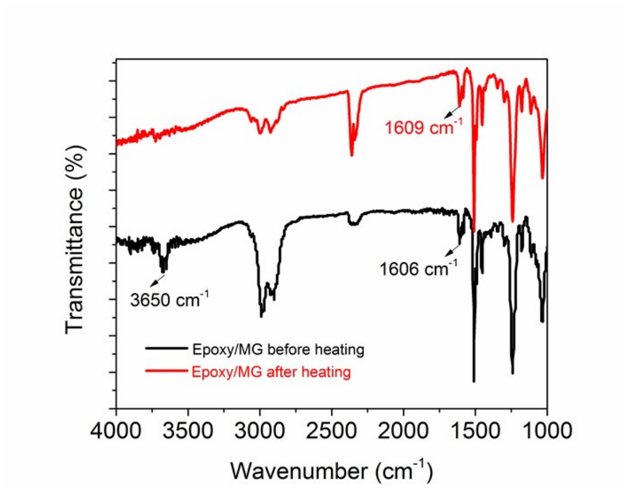
Supplemental Figure S5. SEM of epoxy/MG (3.6 vol%) film surface



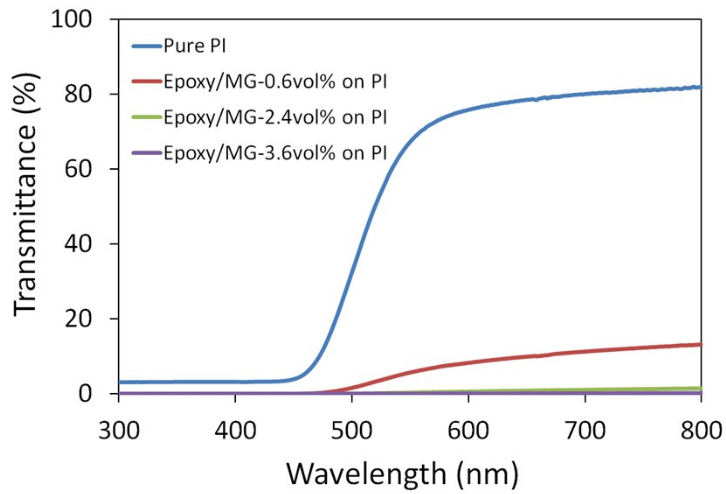
Supplemental Figure S6.Photograph of epoxy/MG (7.5 vol%) film on PI substrate.



Supplemental Figure S7. GISAXS 2D diffractograms of epoxy/MG nanocomposite films at (a) 0.6 vol.%, (b) 2.4 vol.%, (c) 3.6 vol.%, and (d) 5.5 vol.% ZrP. (e) GISAXS 1D diffractogram spectrum of epoxy/MG films at various concentrations.

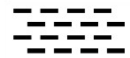
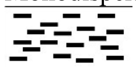
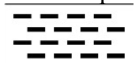
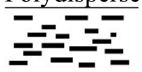
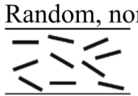


Supplemental Figure S8. FTIR of epoxy/MG before and after heating at 180 ºC for 4hrs. The C=O stretch is observed at 1606 cm^{-1} in the -COOH group on MG surface, and it shifts to 1609 cm^{-1} in the ester group formed by reaction with epoxy.



Supplemental Figure S9. Transmittance spectra of epoxy/MG films at various MG concentrations, illustrating a decreased transparency with increased MG loadings.

Supplemental Table S1 List of permeability models for polymer nanocomposites.

Model	Flake Size, Array Type	Model Dimension	Equation
Nielsen	Monodisperse, Regular Array 	2D	$\frac{P}{P_0} = \frac{1-\phi}{1+\frac{\alpha}{2}\phi}$
Cussler Random Array Monodisperse Flakes	Monodisperse, Random Array 	2D	$\frac{P}{P_0} = \frac{1-\phi}{[1+\frac{\alpha}{3}\phi]^2}$
Cussler Ideal Regular Array	Monodisperse, Regular Array 	2D	$\frac{P}{P_0} = \left(1+\frac{\frac{1}{4}\alpha^2\phi^2}{1-\phi}\right)^{-1}$
Gusev-Lusti	Polydisperse, Random Array 	3D	$\frac{P}{P_0} = e^{-(\frac{\alpha\phi}{3.47})^{0.71}}$
Bharadwaj	Random, non-oriented 	2D	$\frac{P}{P_0} = \frac{1-\phi}{[1+\alpha\phi(2S+1)/6]}$

Note: The Nielsen, Cussler and Bharadwaj models can be viewed as 3D models if the fillers are considered to be ribbons with infinite length. The order parameter S = −1/2, when the ribbons are normal to the membrane; S = 1, when the ribbons are parallel to the membrane surface; S = 0, when the ribbons are randomly oriented.