

Electronic Supplementary Information

Preparation, Fracture, and Fatigue of Exfoliated Graphene

Oxide/Natural Rubber Composites

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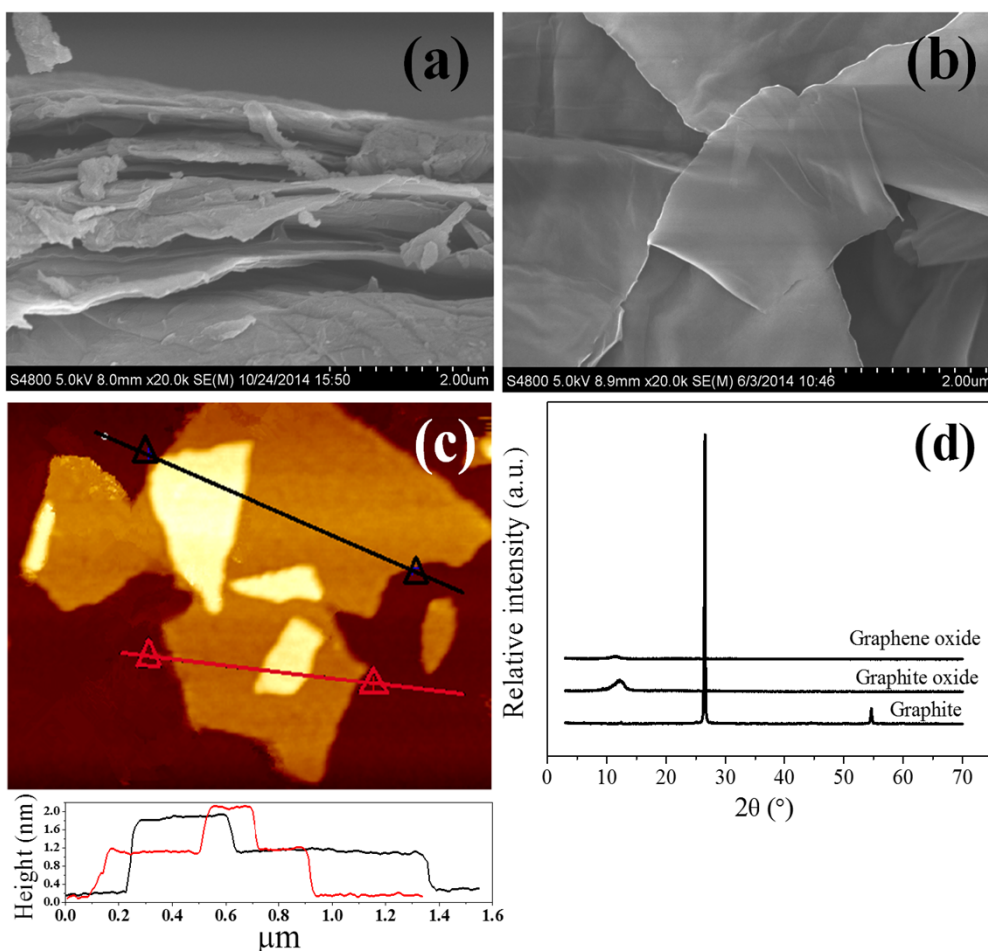


Figure S1. (a) SEM micrograph of frozen-dried graphite oxide; (b) SEM micrograph of frozen-dried exfoliated GO sheets; (c) AFM tapping-mode image and its corresponding height profiles of GO sheets; (d) XRD patterns of pristine graphite, graphite oxide and exfoliated GO sheets.

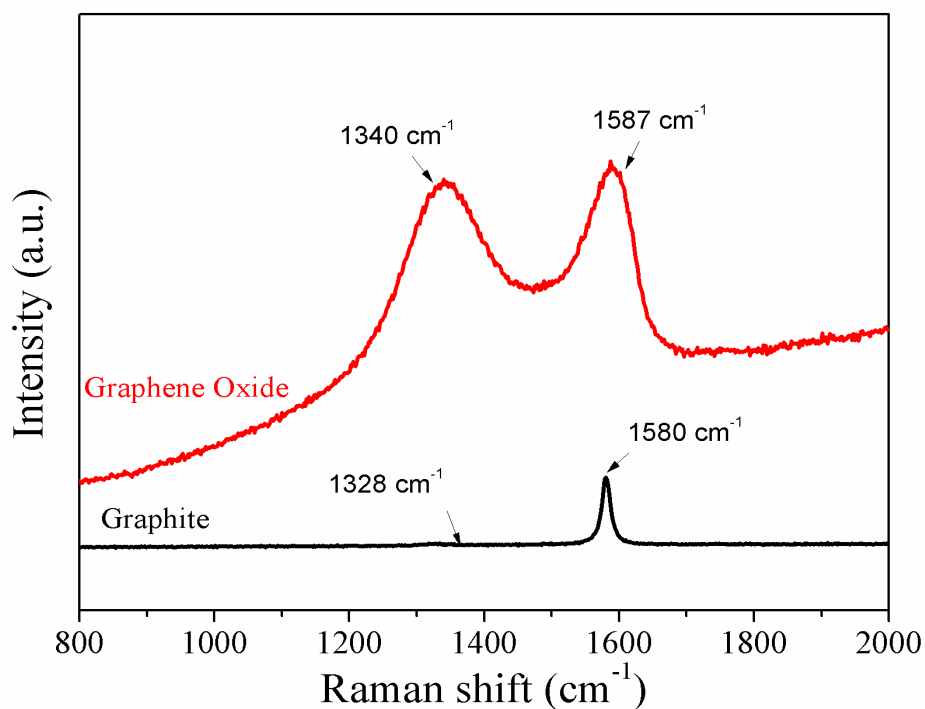
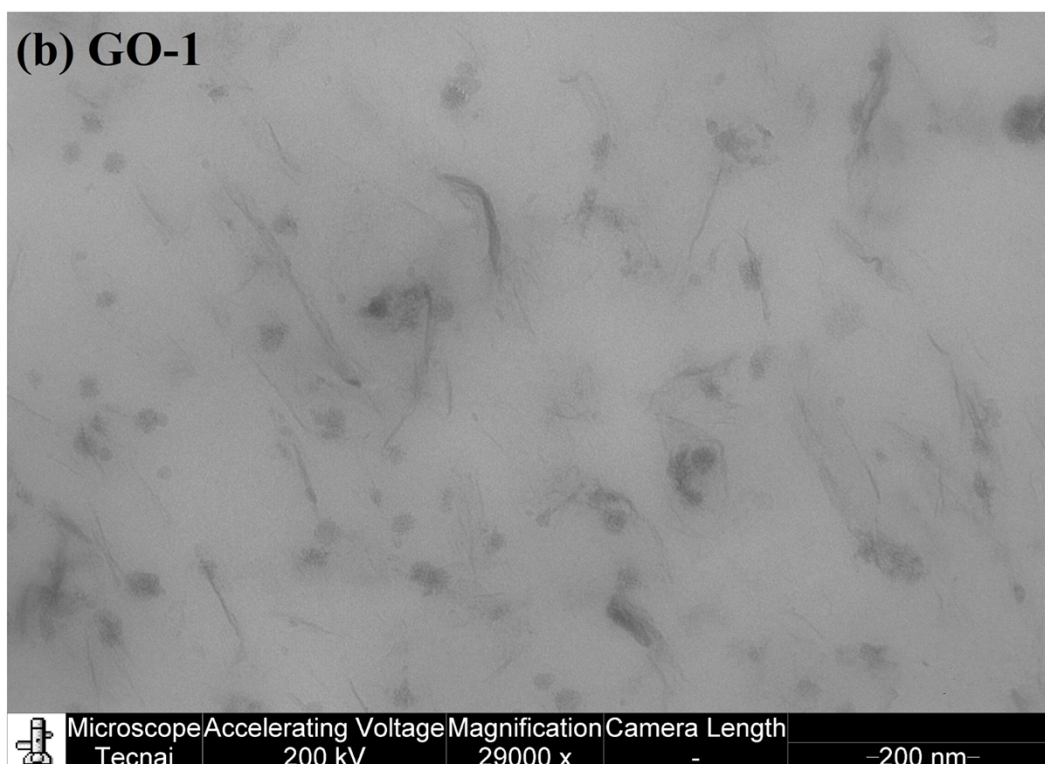
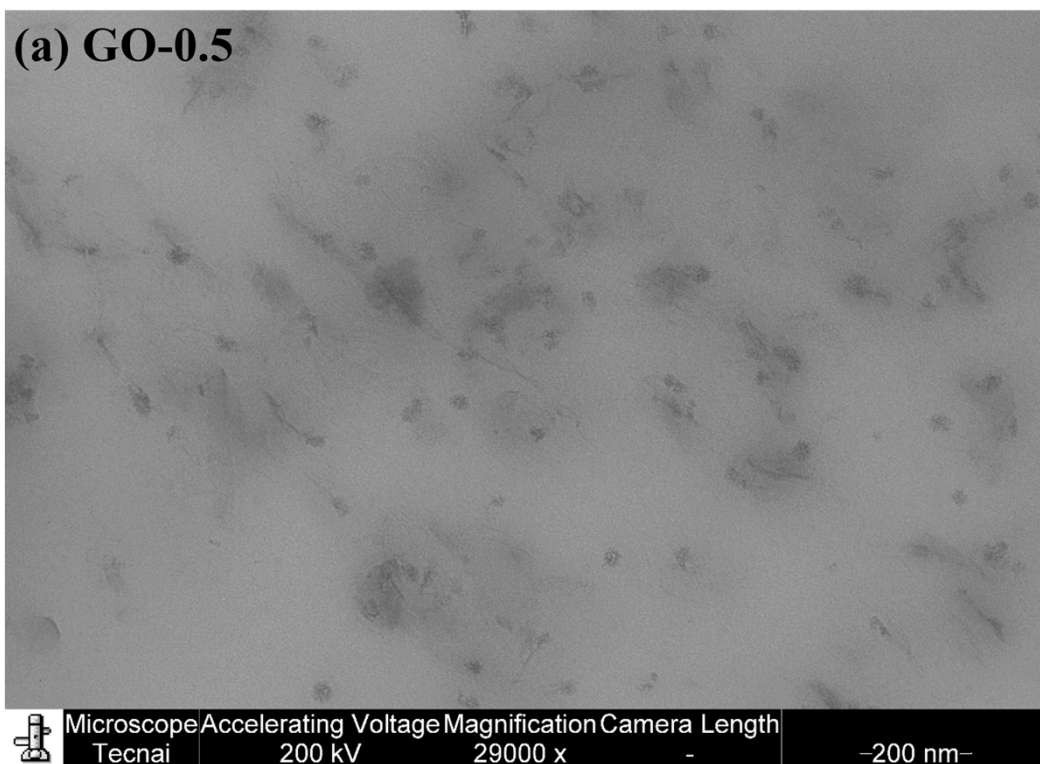


Figure S2. Raman spectra of graphite and graphene oxide.

Raman was used to confirm the changes on the graphite structure of GO due to the oxidation and exfoliation. The pristine graphite contains a D band at 1328 cm⁻¹ and a G band at 1580 cm⁻¹. The GO contains a D band at 1340 cm⁻¹ and a G band at 1587 cm⁻¹. The intensity ratio of D band and G band (I_D/I_G) increased from 0.10 for graphite to 0.91 for GO, suggesting that the ordered structure of graphite was destroyed to some extent after oxidation.



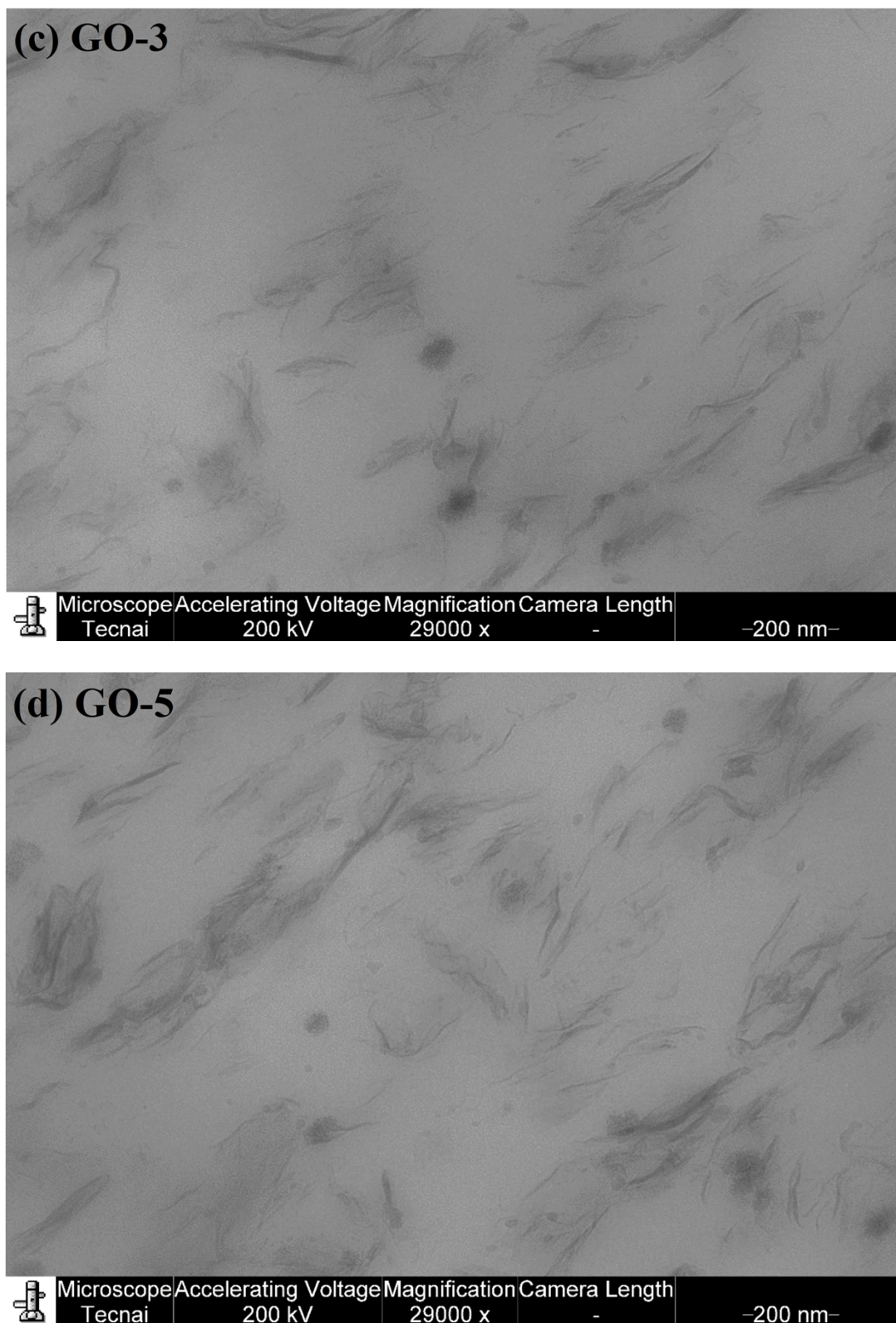


Figure S3. TEM image of GO/NR composites at a higher magnification of 29000:

(a) GO-0.5; (b) GO-1; (c) GO-3; (d) GO-5.

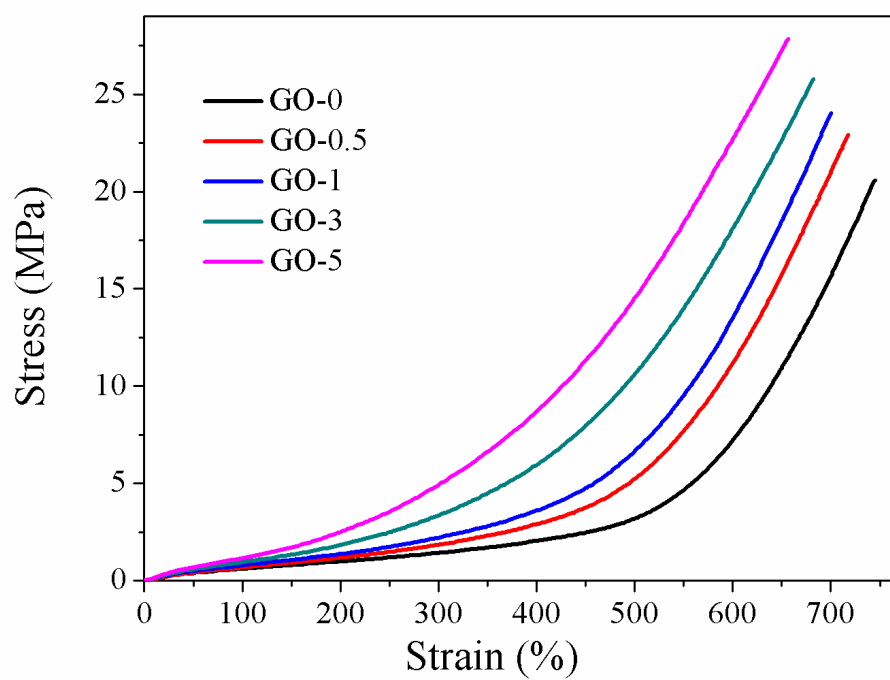


Figure S4. The stress-strain curves of GO/NR composites.

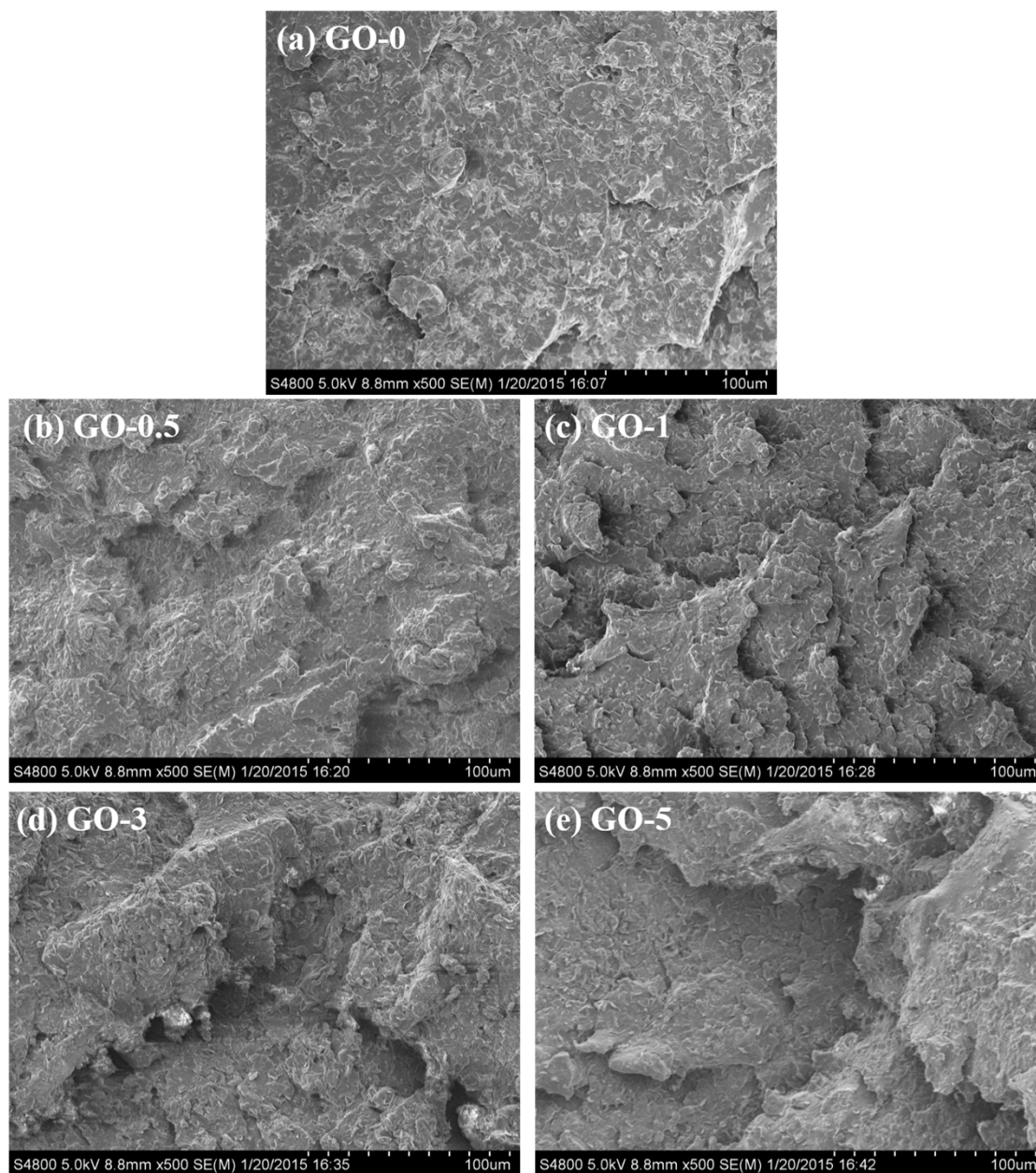


Figure S5. The SEM micrographs of fracture surface after cyclic fatigue: (a) GO-0; (b) GO-0.5; (c) GO-1; (d) GO-3; (e) GO-5.

Table S1. XPS C1s peak position and its relative atomic percentage in different functional groups of graphite and graphene oxide.

Samples	C1s Contribution, at.-%			
	C-C (284.6 eV)	C-O (286.6 eV)	C=O (287.5 eV)	O-C=O (288.5 eV)
Graphite	70.6	20.4	6.8	2.2
Graphene Oxide	42.4	33.0	17.2	7.6

XPS was employed to provide quantitative information about the types and relative amounts of surface oxygen-containing groups of pristine graphite and GO sheets. The C1s XPS spectra were de-convoluted into four different components corresponding to different types of carbon atoms: C-C (284.6 eV), C-O (286.6 eV), C=O (287.5 eV), O-C=O (288.5 eV). The relative percentage contents of functional groups of graphite and GO sheets were summarized in Table S1. The percentage contents of oxygenic groups of GO were highly improved after oxidization.

Table S2. BET surface area of carbon black (N330), silica (VN3), and exfoliated GO sheets.

Samples	Carbon Black (N330)	Silica (VN3)	GO Sheets
BET Surface Area (m ² /g)	81 ¹	175 ²	462 ³

Table S3. Values of C_1 , C_2 and λ_{up} obtained from modified Mooney-Rivlin plots.

Samples	C_1 (MPa)	C_2 (MPa)	λ_{up}
GO-0	0.142	0.066	3.68
GO-0.5	0.162	0.069	3.50
GO-1	0.214	0.078	3.28
GO-3	0.257	0.077	3.02
GO-5	0.294	0.072	2.76

References

- 1 Z. H. Li, J. Zhang and S. J. Chen, *Express Polym. Lett.*, 2008, **2**, 695-704.
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- 3 C. Wan, M. Frydrych and B. Chen, *Soft Matter*, 2011, **7**, 6159-6166.