Electronic Supplementary Material (ESI) for Nanoscale Advances.

Synthesis of Diamine Functionalised Graphene Oxide and Its Application in the Fabrication of Electrically Conducting Reduced Graphene Oxide/Polymer Nanocomposite Films

Namrata Maslekar,[1] Rabiatul A.M. Noor,[1] Rhiannon P. Kuchel,[2] Yin Yao,[2] Per B. Zetterlund,[1,*] Vipul Agarwal[1,*]

[1]*Centre for Advanced Macromolecular Design (CAMD), School of Chemical Engineering, University of New South Wales, Sydney, NSW 2052, Australia*
[2]*Mark Wainwright Analytical Centre, University of New South Wales, Sydney, NSW, 2052, Australia*
* Corresponding authors: agarwalvipul84@gmail.com, p.zetterlund@unsw.edu.au

Supporting Information

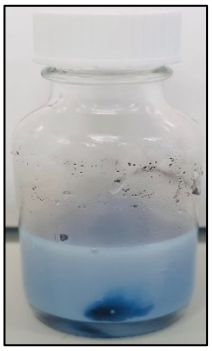
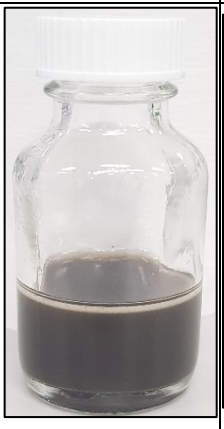
Table S1: Experimental details of EDA functionalisation of GO (GO-NH_2) and corresponding XPS analysis.

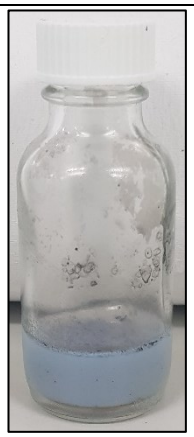
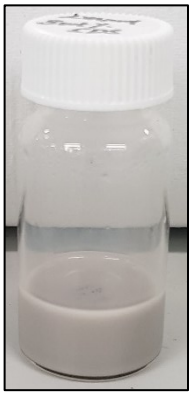
GO (0.4 wt%) (mL)	EDA (mM)	EDC (g)	NHS (g)	Reaction time (h)	XPS Elemental at%		
					C	O	N
2	12	0.4	0.2	24	66.48	26.24	7.29
2	24	0.4	0.2	24	66.20	26.50	7.30
2	6	0.4	0.2	24	65.28	28.46	6.25
2	4	0.2	0.1	24	66.30	25.60	8.04
2	12	-	-	24	67.97	26.88	5.15
2	2	-	-	24	65.55	27.02	5.36
2	1	-	-	24	65.23	29.34	4.68
25	0.125	-	-	0.5	69.46	25.01	1.21
1	-	-	-	-	64.31	33.86	-

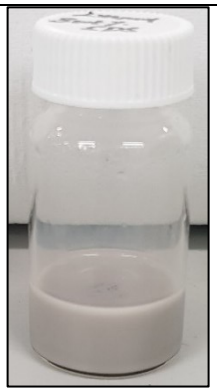
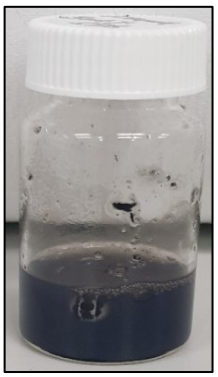
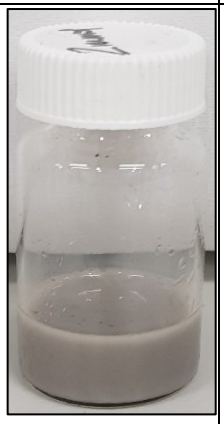
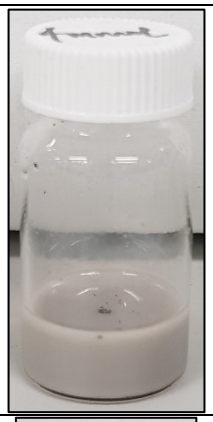
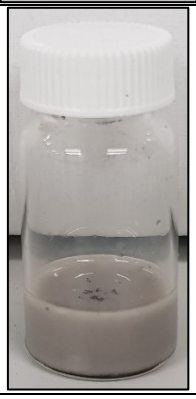
Table S2: Deconvoluted C1s peak descriptions for both GO and as-synthesised GO-NH_2 (1.2 at% nitrogen) used in the polymerisation reactions

Functionality	GO	GO-NH_2
	Binding energy (eV) (atom%)	Binding energy (eV) (atom%)
C-C/C=C	284.80 (30.64%)	284.80 (38.48%)
C-N/O-H	-	285.60 (6.85%)
C-O-C	286.87 (26.23%)	287.05 (7.35%)
C=O	287.52 (6.06%)	287.54 (13.99%)
COOH	288.85 (2.8%)	289.04 (2.79%)

Table S3: GO-NH_2 synthesis reaction conditions and miniemulsion polymerisation under different reaction conditions (AIBN and HD concentrations remained the same in all reactions), and images of the miniemulsion (before polymerisation) and polymer latex (after polymerisation). Clear phase separation pertains to the emergence of clear white (monomers/polymer) and dark layers (GO).

Serial number	**GO-NH_2 synthesis condition**			**Polymerisation conditions**			**Before Polymerisation**	**After Polymerisation**
	GO (mL)	EDA (mM)	Reaction duration (h)	GO-NH_2 (wt%)	Monomers (wt%)	SDS (wt%)		
1.	25	1	0.5	5	7	3		
2.	25	0.125	0.5	1	7	1		
3.	25	0.125	0.5	5	7	1		

4.	25	0.375	2	3	5	3		
5.	25	0.375	4	5	5	3		
6.	2	2	24	1	5	3		
7.	2	6	24	1	5	1		-

8.	2	24	24	1	5	1		
9.	2	2	24	1	5	1		
10.	2	4	24	1	5	1		
11.	2	6	24	1	5	1		

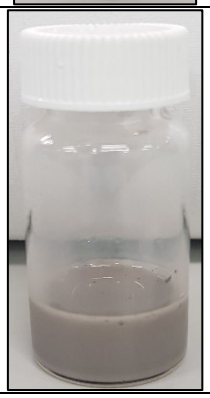
12.	2	8	24	1	5	1		
13.	2	12	24	1	5	1		
14.	2	24	24	1	5	1		

Table S4: Miniemulsion polymerization recipes of St/nBA at 70 °C using GO-NH_2 (1.2 atom% nitrogen). (Fixed amounts of AIBN, monomers and hexadecane at 0.25 M (relative to organic phase), 7 wt% (relative to water), and 5 wt% (relative to monomer), respectively).

Serial Number	**GO-NH_2 (wt%)**	**SDS (wt%)**	**Polymerization**	**Conversion (%)**
1.	5	1	Phase Separation	-
2.	5	2	Phase Separation	-
3.	5	3	Yes	68
4.	5	5	Yes	74
5.	1	1	Phase Separation	-

6.	3	1	Phase Separation	-
7.	3	2	Phase Separation	-

Table S5: Particle size (DLS) data, number-average (M_n) and weight-average (M_w) molecular weights and dispersity (Đ) for St/nBA/GO-NH_2 (5 wt%) polymers at different concentrations of SDS. *Poor autocorrelation fit (data included as rough indication).

SDS (wt%)	**Particle diameter (nm)**	**PDI**	**M_n (x 10^4) (kg/mol)**	**M_w (x 10^5) (kg/mol)**	**Đ**
3	6890	1.00*	12.45	10.25	8.2
5	5470	0.97*	12.3	9.9	8.1

Table S6: Water contact angle on different unreduced and thermally reduced St/nBA/GO-NH_2 (5 wt%) nanocomposite films at different concentrations of SDS. Data is presented as average ± standard deviation (SD) calculated from the contact angle measured from 2 individual films and 2 measurements per film

Sample	**Contact angle (°)**
St/nBA/GO-NH_2/ SDS (3 wt%)	65±3
St/nBA/GO-NH_2/ SDS (5 wt%)	60±2
St/nBA/rGO-NH_2/ SDS (3 wt%)	84±2
St/nBA/rGO-NH_2/ SDS (5 wt%)	82±2

Table S7: Surface roughness (R_a) of unreduced and thermally reduced St/nBA/GO-NH_2 (5 wt%) nanocomposite films at different concentrations of SDS. Data is representative of 2 individual films and 3 measurements per film and presented as average ± standard deviation (SD)

Sample	**Surface roughness (µm)**
St/nBA/GO-NH_2/ SDS (3 wt%)	0.97 ± 0.10
St/nBA/GO-NH_2/ SDS (5 wt%)	1.14 ± 0.10
St/nBA/rGO-NH_2/ SDS (3 wt%)	1.15 ± 0.06
St/nBA/rGO-NH_2/ SDS (5 wt%)	1.09 ± 0.09

Table S8: Electrical conductivity values obtained for thermally reduced control and St/nBA/GO-NH_2 (5 wt%) nanocomposite films at different concentrations of SDS. Data is representative of 2 individual films and 3 measurements per film and presented as average ± standard deviation (SD)

Sample	**Electrical conductivity (S m^{-1})**	**Reference**
St/nBA/rGO-NH_2/ SDS (3 wt%)	0.79 ± 0.0001	This work
St/nBA/rGO-NH_2/ SDS (5 wt%)	0.89 ± 0.0370	This work

St/nBA/rGO/ SDS (1 wt%)	2.49± 0.15	[1]

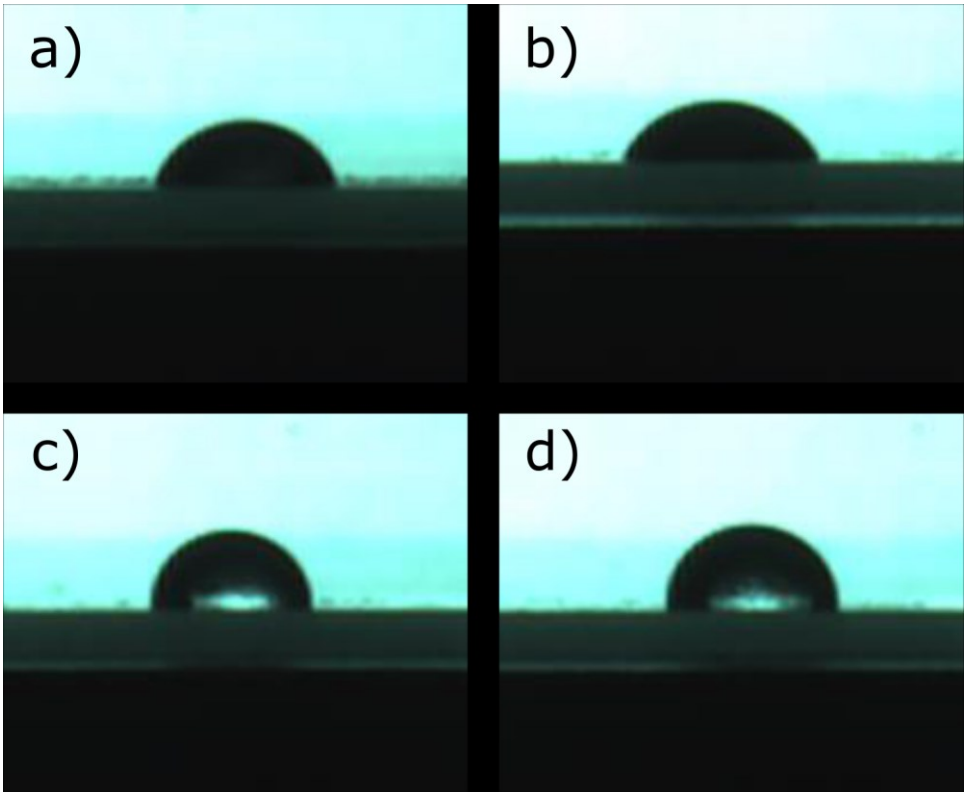


Figure S1: Water contact angle images of the St/nBA/GO-NH2 (5%) nanocomposite films with 3 and 5 wt% SDS (a and b) before and (c and d) after thermal reduction. The obtained images were analysed using ImageJ.

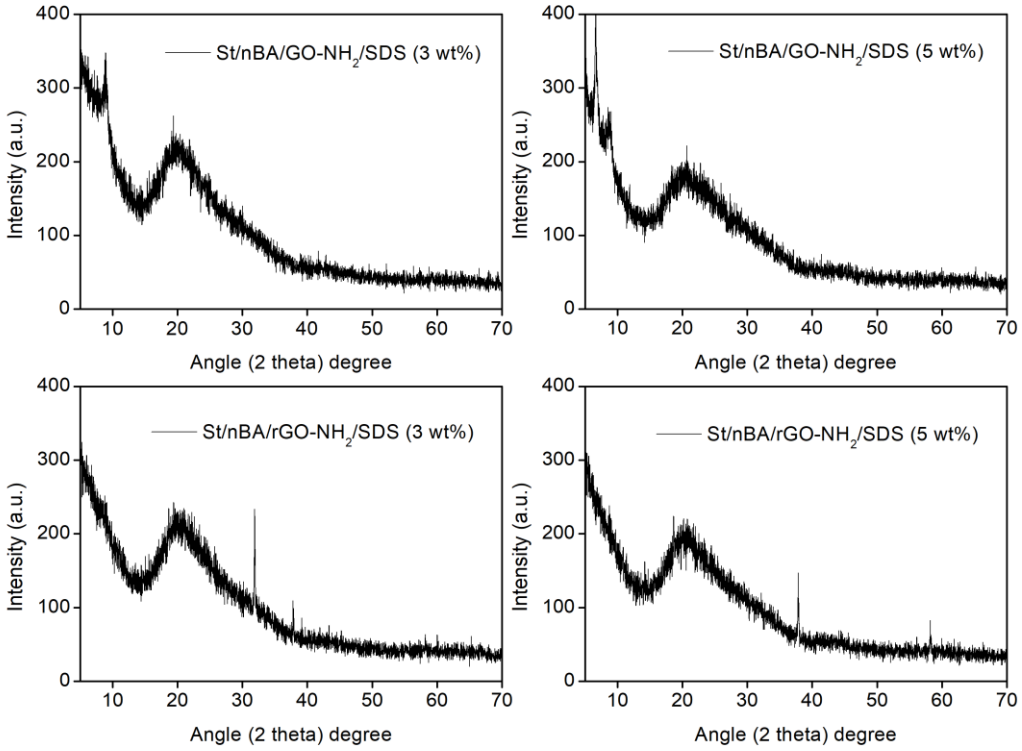


Figure S2: XRD of the St/nBA/GO-NH2 (5%) nanocomposite films with 3 and 5 wt% SDS before and after thermal reduction. The peak at ~9° in the case of unreduced St/nBA/GO-NH_2 (5%) nanocomposite films (top row) can be attributed to (001) plane of GO while the broad peak centred ~20° is the contribution of the polymer.[2, 3] The loss of (001) peak in the reduced nanocomposite films (bottom row) confirms the successful reduction of the GO to rGO within the nanocomposite films.

References

1. Y. Fadil, L. N. M. Dinh, M. O. Y. Yap, R. P. Kuchel, Y. Yao, T. Omura, U. A. Aregueta-Robles, N. Song, S. Huang, F. Jasinski, S. C. Thickett, H. Minami, V. Agarwal and P. B. Zetterlund, *ACS Appl. Mater. Interfaces*, 2019, **11**, 48450-48458.
2. V. Abhilash, N. Rajender and K. Suresh, in *Spectroscopy of Polymer Nanocomposites*, Elsevier, 2016, pp. 410-451.
3. Y. Cai, Y. Fadil, F. Jasinski, S. C. Thickett, V. Agarwal and P. B. Zetterlund, *Carbon*, 2019, **149**, 445-451.