Supporting Information

Graphene-Oxide Functionalized with 2-Ureido-4[1*H*]-pyrimidinone for Production of Nacre-Like Films

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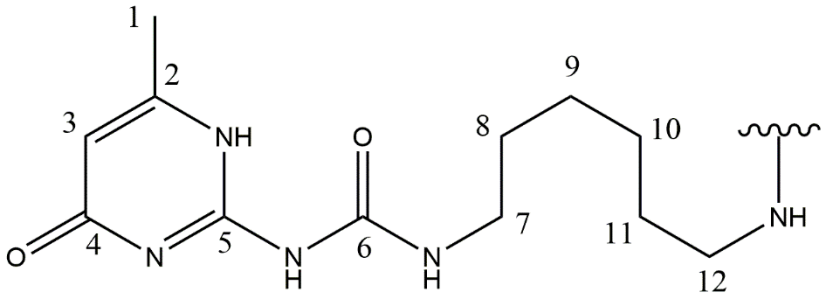
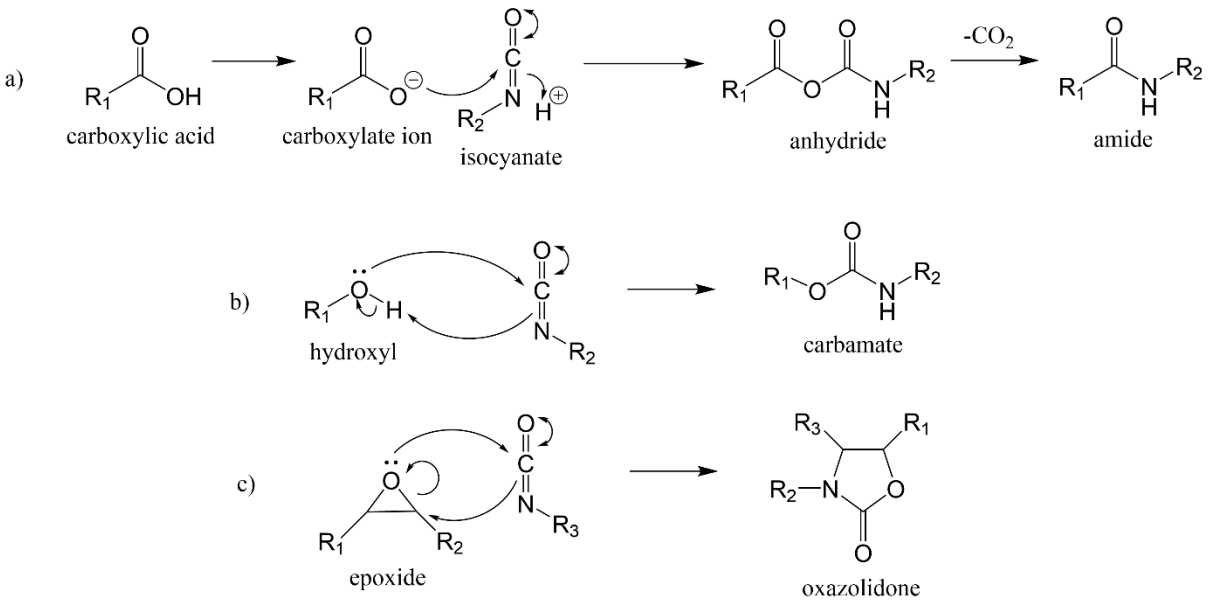


Figure S1. Chemical structure of the UPy isocyanate when bound to GO.



Scheme S1. a) Reaction between a carboxylic acid group and isocyanate to produce an amide via a condensation reaction, b) reaction between a hydroxyl group and an isocyanate to produce a carbamate and c) reaction between an epoxide and an isocyanate to produce an oxazolidone.

Table S1. Elemental composition and carbon to nitrogen bonding composition for GO and GO.UPy samples as determined from XPS.

	Elemental Composition			**N_{1s} region (carbon-nitrogen bonding)**		
Sample	C (%)	O (%)	N (%)	C=N (%)	C-N (%)	C-N corrected (%)
GO	65.84	32.76	0	0	0	0
GO.UPy.99.1	67.09	30.55	0.61	0	28.24	28.24
GO.UPy.90.10	67.90	28.54	1.68	0	46.04	46.04
GO.UPy.50.50	66.78	23.73	7.68	15.15	57.92	50.41

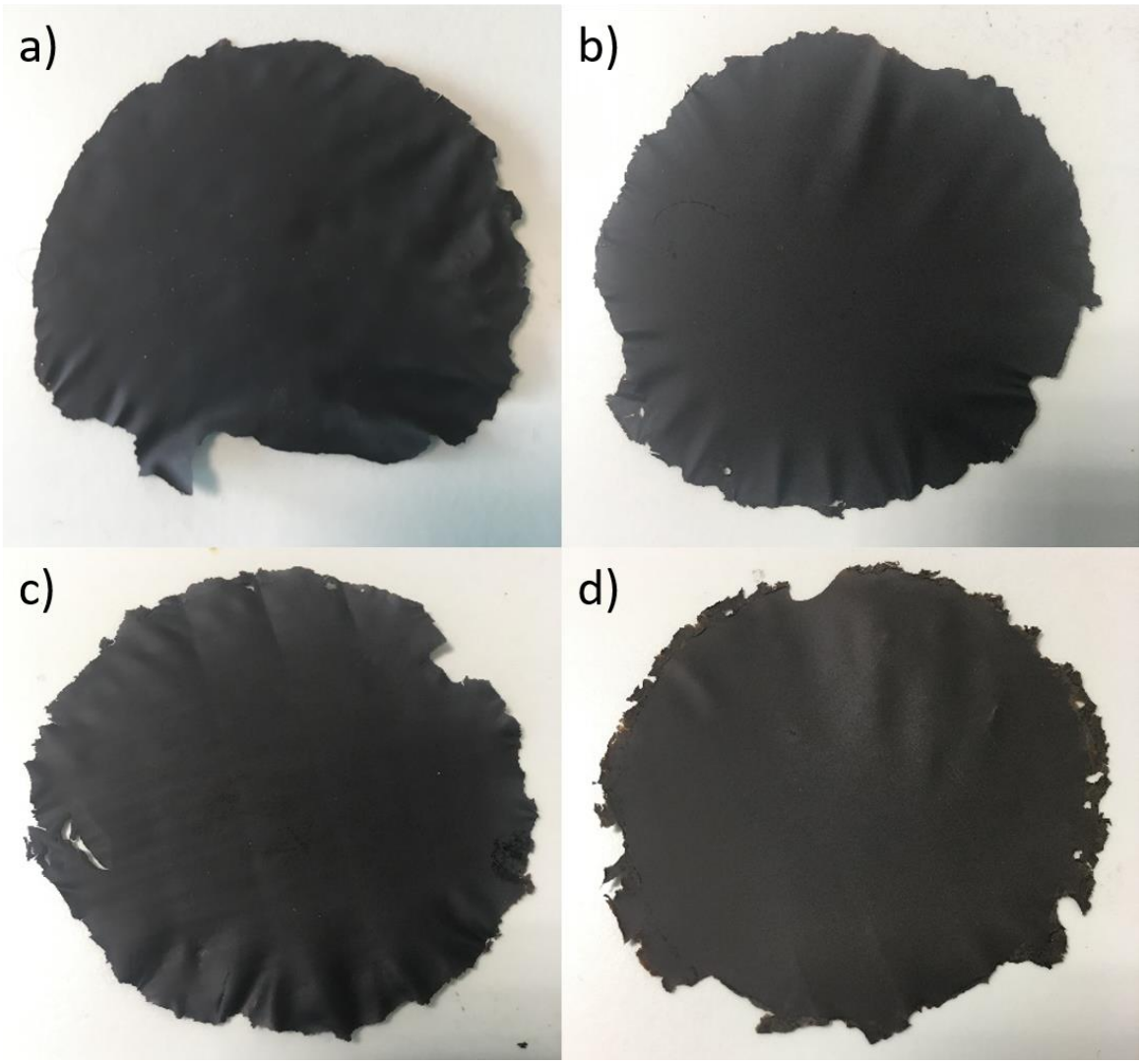


Figure S2. Digital photographs of the GO and GO.UPy films a) neat GO b) GO.UPy.99.1, c) GO.UPy.90.10 and d) GO.UPy.50.50.

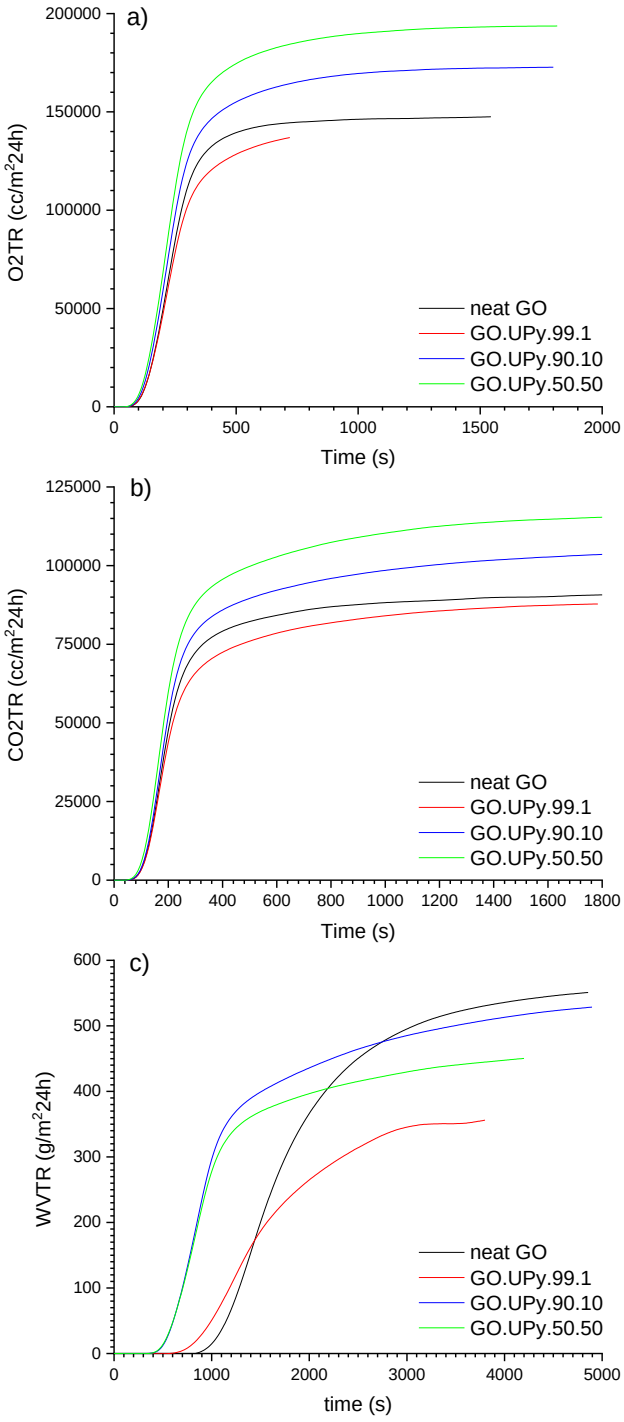


Figure S3. Gas barrier transmission curves for GO and GO.UPy samples, a) oxygen transmission, b) CO_2 transmission and c) water vapour transmission rate (WVTR).

Table S2. Dispersions of GO.UPy.99.1 in a range of solvents displaying stability for up to 96 hours.

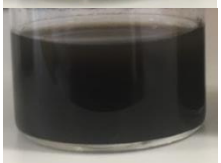
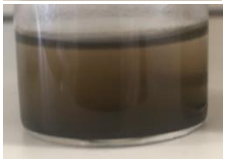
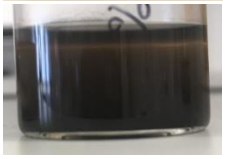
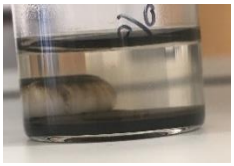
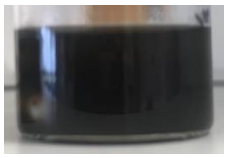
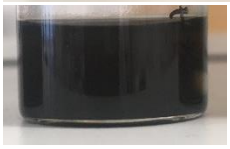
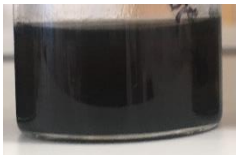
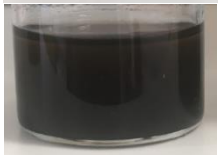
Time (h)	0	1	24	96
Acetone				
Ethylacetate				
Isopropanol				
DMF				
DSMO				
DCM				
THF				

Table S3. ^{13}C MAS resonances, intensities and assignments for GO and GO.UPy samples. Assignment numbers correlate to the chemical structure in figure S1 and functional groups in brackets correspond to adjacent group connecting the UPy to the GO in the case of carbon #12.

GO neat δ_{iso} (ppm)	I (%)	GO.UPy.99.1 δ_{iso} (ppm)	I (%)	GO.UPy.90.10 δ_{iso} (ppm)	I (%)	GO.UPy.50.50 δ_{iso} (ppm)	I (%)	Assignment
						19.1	0.9	1
						30.0	1.8	12 (amide)
		31.8	4.5	31.8	3.5	31.8	2.4	8, 11
						35.5	3.2	7
		37.5	4.2	37.5	3.4	39.1	2.8	9, 10
						44.6	1.3	12 (carbamate)
61.8	13.4	60.9	12.0	60.9	6.1	60.9	4.1	C-O-C
70.2	7.0	71.0	7.4	71.0	5.9	71.0	3.7	C-O-H
121.4	57.8	125.1	56.7	118.9	59.2	129.5	69.1	sp^2, 2, 5
131.7	16.5	132.0	9.7	132.0	14.9	132.0	3.2	sp^2
						150.5	0.3	3
						156.4	0.9	6
166.1	5.3	165.4	5.5	165.9	7.1	166.1	5.7	COOH
						174.7	0.4	4

Table S4. Average values (±SD) for a) Young's modulus, b) tensile strength, c) strain and d) tensile toughness for nacre-like GO and GO.UPy films.

	Young's Modulus (GPa)	**Tensile Strength (MPa)**	**Maximum Strain (%)**	**Tensile Toughness (kJ/m^3)**
GO neat	2.75 ± 0.44	6.45 ± 0.97	0.28 ± 0.06	0.89 ± 0.31
GO.UPy.99.1	3.12 ± 0.53	11.74 ± 1.62	0.37 ± 0.12	2.63 ± 0.73
GO.UPy.90.10	7.35 ± 1.88	23.75 ± 1.13	0.40 ± 0.07	5.23 ± 0.60
GO.UPy.50.50	8.87 ± 1.95	30.43 ± 7.37	0.60 ± 0.06	9.94 ± 3.99