Supporting Information

Self-Assembly of Graphene Oxide Flakes for Smart and Multifunctional Coating with Reversible Formation of Wrinkling Patterns

Xinran Su,[‡]a Rakesh K Pandey, ^{‡a,b} Junhao Ma,^a Wei Chun Lim,^a Chi Kit Ao,^a Changhui Liu,^a Hideyuki Nakanishi,^{*b} Siowling Soh,^{*a}

a. Department of Chemical and Biomolecular Engineering, National University of Singapore, 4 Engineering Drive 4, Singapore 117585, Singapore.

b. Department of Macromolecular Science and Engineering, Graduate School of Science and Technology, Kyoto Institute of Technology, Matsugasaki, Kyoto 606-8585, Japan.



Fig.S1 Analysis of the surface topology of the graphene oxide (GO) coating on a piece of glass substrate by a profilometer. The scan first started on the bare uncoated glass substrate before it reached the coated region. It then continued through the coated region until it reached the opposite side of the bare uncoated glass substrate. The testing parameters included the scan speed of 50 μ m s⁻¹, sampling rate of 50 Hz, and stylus force of 2.54 mg. The average thickness of the GO coating was calculated to be 3.9 μ m.



Fig.S2 Characterizations of GO film (wet) without glass slide. Profilometer testing of the GO film (wet) on PTFE substrate. The scan first started on the bare PTFE substrate before it reached the GO film region. It then continued through the GO film region until it reached the opposite side of the bare PTFE substrate. The testing parameters included the scan speed of 50 μ m s⁻¹, sampling rate of 50 Hz, and stylus force of 2.54 mg. The average thickness of the GO coating was calculated to be 26.6 μ m, and the roughness was 29.3 μ m.



Fig. S3 Optical images of GO coatings prepared from solutions containing different concentrations of GO flakes: (A) 2.7 wt.%, (B) 6.2 wt.%, (C) 7.1 wt.%, (D) 8 wt.%, (E) 8.9 wt.%, and (F) 9.8 wt.%.



Fig. S4 Wrinkles did not form after placing a drop of water on a thick GO coating. GO coatings prepared with two layers (A1 and A2) and three layers (B1 and B2) of coating. Two layers of coating had a thickness of \sim 8 µm and three layers of coating had a thickness of \sim 12 µm. Images show before (A1 and B1) and after (A2 and B2) placing a drop of water on the coatings

Sample	Thickness (µm)	Roughness (µm)					
During preparation of the coating							
Higher temperature	46.0	48.9					
Lower temperature	5.8	7.9					
Agitation	8.9	12.4					
After formation of the coating							
Pressed	2.1	3.1					
Heated	6.8	9.4					
Solvent applied	7.1	9.6					

 Table S1 Analysis by profilometer of samples prepared under different conditions.

Concentrations of GO flakes	6.2 wt.%	7.1 wt.%	8 wt.%	8.9 wt.%	9.8 wt.%
Diameter of wrinkle (mm)	9.3	9.8	11.8	No wrinkle	No wrinkle
Time of wrinkle formation (s)	42	53	46	/	/
Time of wrinkle disappearance (s)	135	137	127	/	/

Table S2 Characteristics of the wrinkles formed by GO coatings prepared from solutions containing different concentrations of GO flakes

Table S3 Characteristics of the GO coatings prepared at different temperatures						
Temperature (°C)	25	35	50	60	75	
Diameter of wrinkle (mm)	11.8	11.1	8.9	7.8	No wrinkle	
Time of wrinkle formation (s)	46	32	30	26	/	
Time of wrinkle disappearance (s)	127	86	38	34	/	