

Negatively-charged lipids exhibit negligible effects on the water repellency of montmorillonite films

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S1. Wettability data. All data points for the measured contact angle are presented in **Figure S1**. Note that there could be two populations of contact angle in the montmorillonite film, one at 42° and one at 34°. The 42° population does not exist in the samples with lipid but the 35° population persists. Average wettability data points for each concentration of lipid are presented in **Table S1**.

Sample	Lipid Concentration (% CAM)	Contact angle error (°)	Contact angle error (°)	Time constant (s)	Time constant error (s)	Average α	α error
Montmorillonite	0	37.3	3.5	2.0	0.1	0.016	0.004
DOPG	10	33.6	2.5	2.1	0.2	0.016	0.003
	20	33.3	2.9	2.2	0.2	0.015	0.004
	25	35.7	2.2	2.4	0.2	0.019	0.002
	30	32.6	1.7	2.4	0.2	0.016	0.002
	40	32.3	2.8	2.3	0.2	0.015	0.002
	50	30.3	2.5	2.3	0.2	0.015	0.003
DSPG	10	34.3	2.0	2.1	0.1	0.015	0.002
	20	34.0	1.8	2.3	0.2	0.016	0.001
	25	34.2	1.7	2.2	0.2	0.015	0.003
	30	33.3	1.2	2.1	0.2	0.014	0.004
	40	34.7	1.8	2.4	0.1	0.016	0.005
	50	35.3	1.0	2.3	0.1	0.018	0.003

S2. Addendum to 3.1.1 Lipid Aggregate Behavior. Overall, the fraction of aggregates on edges is always under 30% or over 70%. We speculate that this may be due to aggregate size determining how easily the aggregates could slide between montmorillonite flakes, but plotting the aggregate size versus the edge fraction does not yield any obvious trends (**Figure S12**).

Aggregate heights measured for the DSPG 25% sample are provided in **Figure S2B**. Additional height profiles for the bilayer sheet present in the DSPG 50% sample are provided in **Figure S3**.

S3. TC vs. α . PG wettability variables are plotted on an expanded axis in **Figure S4**. For PG lipids, there is no relationship between α and TC (**Figure S5**).

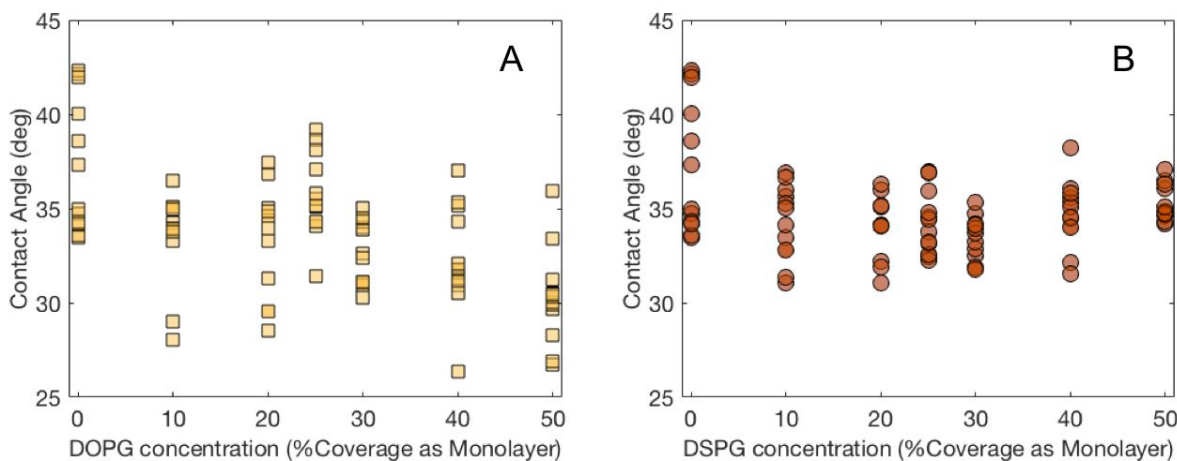


Figure S1: All contact angle data points for A) DOPG and B) DSPG. The points are transparent, so the color intensity is higher when the points overlap.

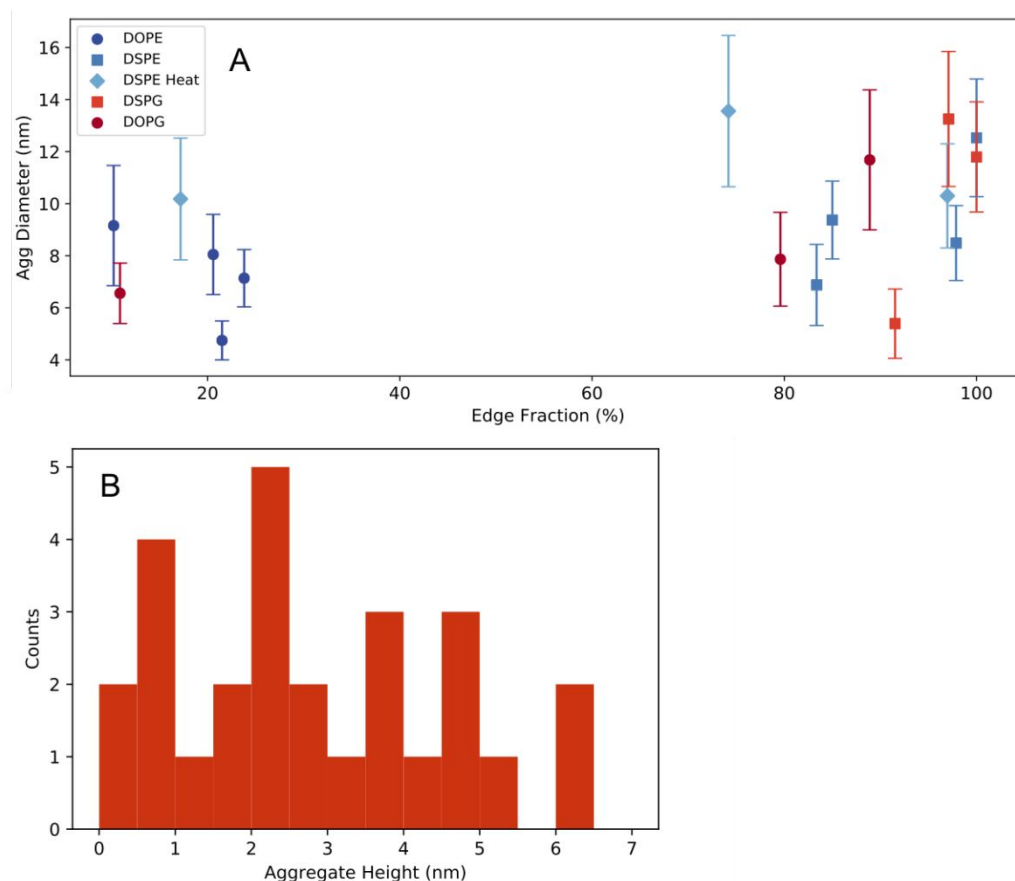


Figure S2: Aggregate characteristics. (A) Aggregate diameter and the fraction of aggregates on clay edge sites are unrelated. (B) Aggregate heights for DSPG at 25% coverage. Mean = 2.8 nm, standard deviation = 1.7 nm. Each bin is 0.5 nm.

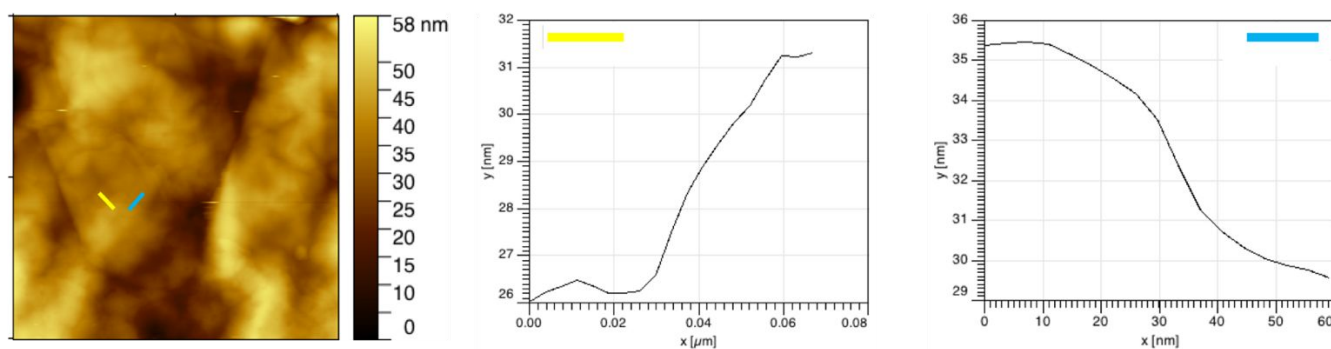


Figure S3: Additional height profiles for the bilayer seen in the DSPG 50% concentration.

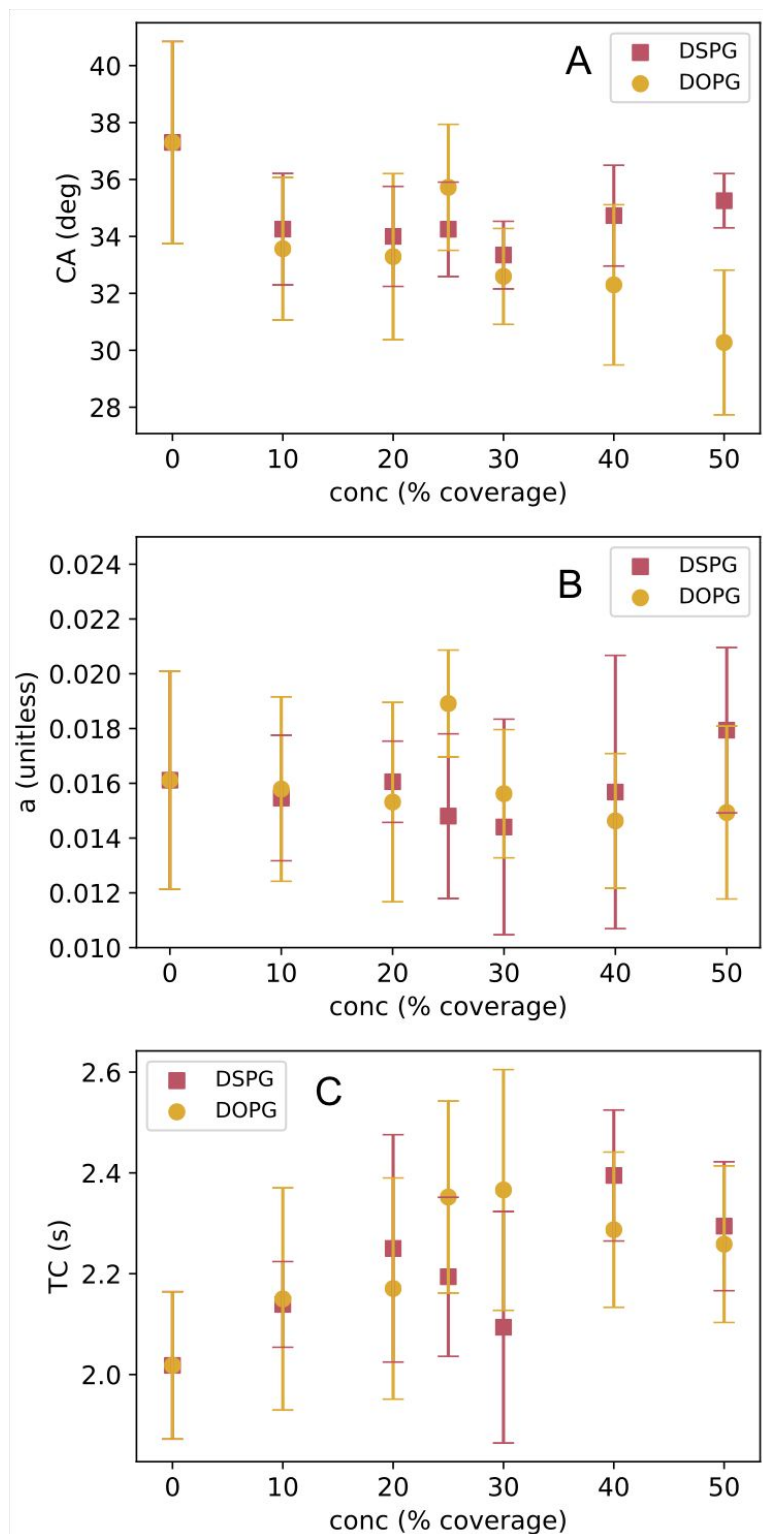


Figure S4: Wettability variables vs. concentration for PG lipids. (A) Contact angle against concentration. (B) Exponent a against concentration. (C) The time constant TC (or $1/K$ in eqn. 2) against concentration. Error bars denote standard deviations.

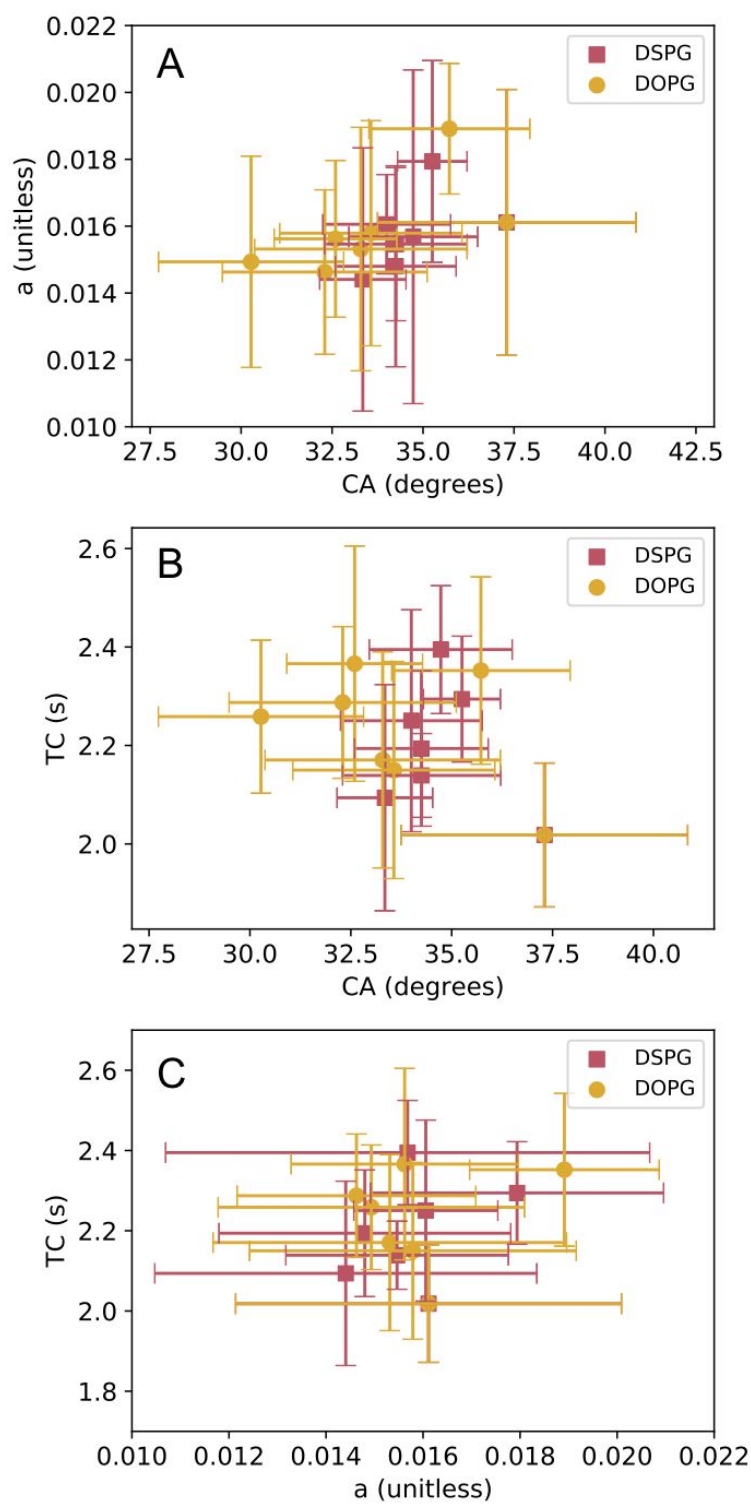


Figure S5: (A) a vs TC for PG lipids. (B) TC vs. CA for PG lipids. (C) CA vs a for PG lipids.

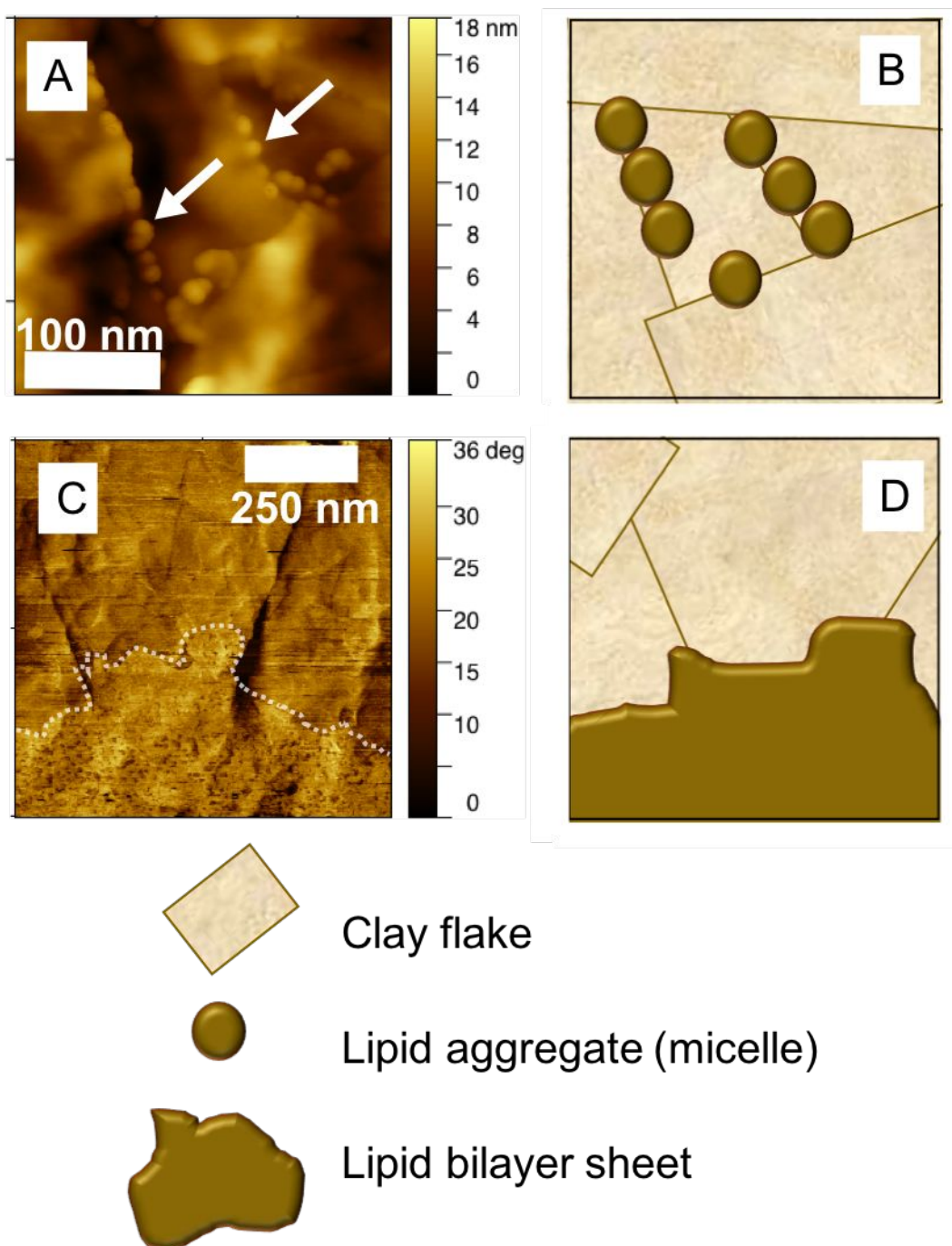


Figure S6: A) AFM topography showing aggregates on the edges of clay flakes. B) A cartoon of the image in (A). C) AFM phase image showing the presence of a lipid bilayer sheet on the clay surface. D) A cartoon of (C).

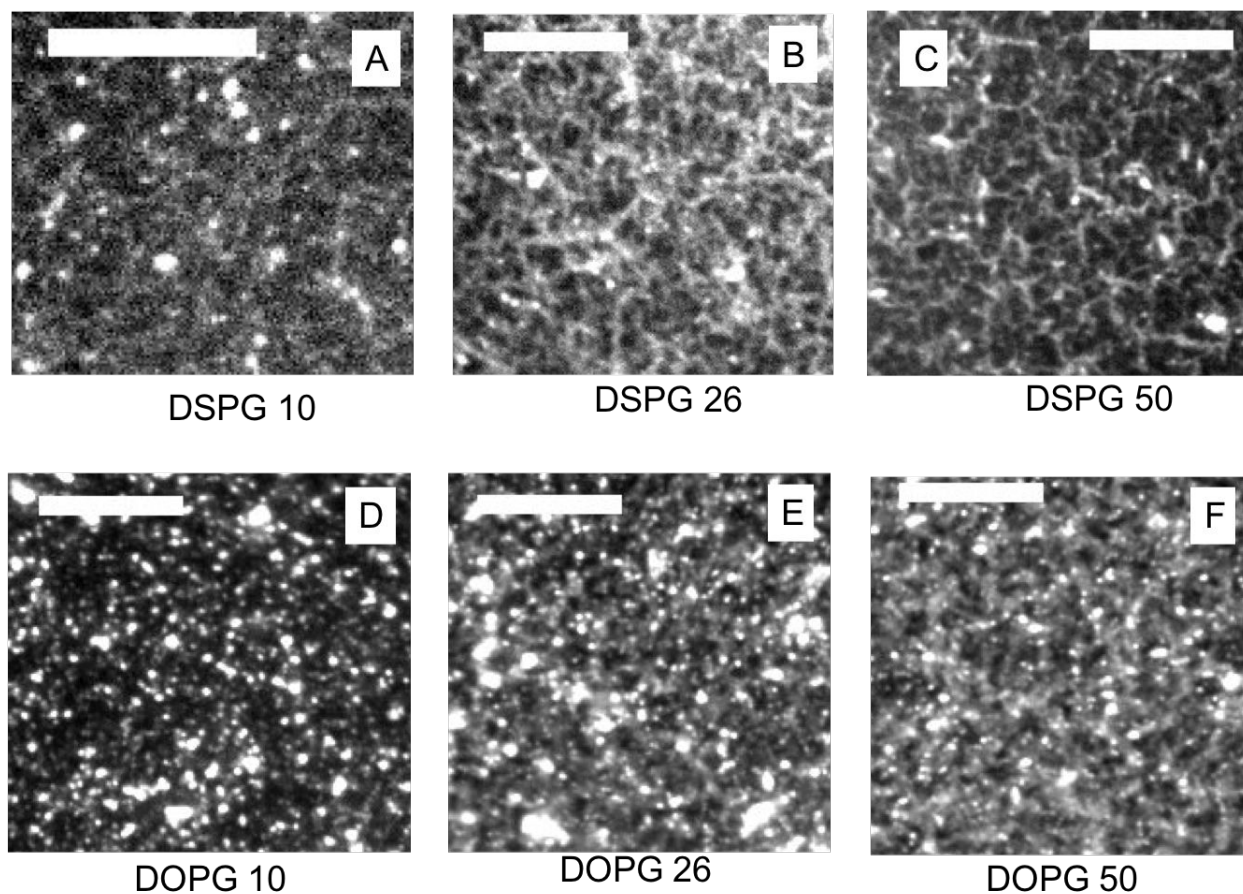


Figure S7: Representative fluorescence micrographs for several coverage values. Scale bars are 50 μm . A) DSPG 10% coverage. B) DSPG 26% coverage. C) DSPG 50% coverage. D) DOPG 10% coverage. E) DOPG 26% coverage. F) DOPG 50% coverage.

Table S2. r^2 values for each linear comparison of wetting and physical variables. Larger values are indicated by a darker color. CA = Contact angle

	DOPG			DSPG		
	CA	TC	α	CA	TC	α
Avg diameter	0.13	0.56	0.00	0.97	0.85	0.44
Aggregate/nm ² estimate	0.35	0.30	0.04	0.99	0.79	0.51
edge/total aggregates	0.76	0.02	0.34	0.89	0.36	0.90
Fluorescence 25% Threshold, Junctions	0.20	0.46	0.00	0.07	0.35	0.00
Fluorescence 50% Threshold, Junctions	0.43	0.23	0.07	0.45	0.13	0.64
Fluorescence 75% Threshold, Junctions	0.38	0.27	0.05	0.72	0.96	0.53
Fluorescence 25% Threshold, Skeleton ratio	0.09	0.62	0.02	0.52	0.85	0.33
Fluorescence 50% Threshold, Skeleton ratio	0.18	0.48	0.00	0.48	0.81	0.29
Fluorescence 75% Threshold, Skeleton ratio	0.31	0.34	0.02	0.51	0.84	0.32
AFM 25% Threshold, Junctions	0.06	0.67	0.04	0.36	0.71	0.19
AFM 50% Threshold, Junctions	0.01	0.80	0.11	0.25	0.60	0.10
AFM 75% Threshold, Junctions	0.31	0.34	0.02	0.29	0.64	0.13
AFM 25% Threshold, Skeleton ratio	0.38	0.27	0.05	0.39	0.74	0.21
AFM 50% Threshold, Skeleton ratio	0.70	0.65	0.98	0.29	0.64	0.13
AFM 75% Threshold, Skeleton ratio	0.14	0.54	0.002	0.30	0.65	0.14

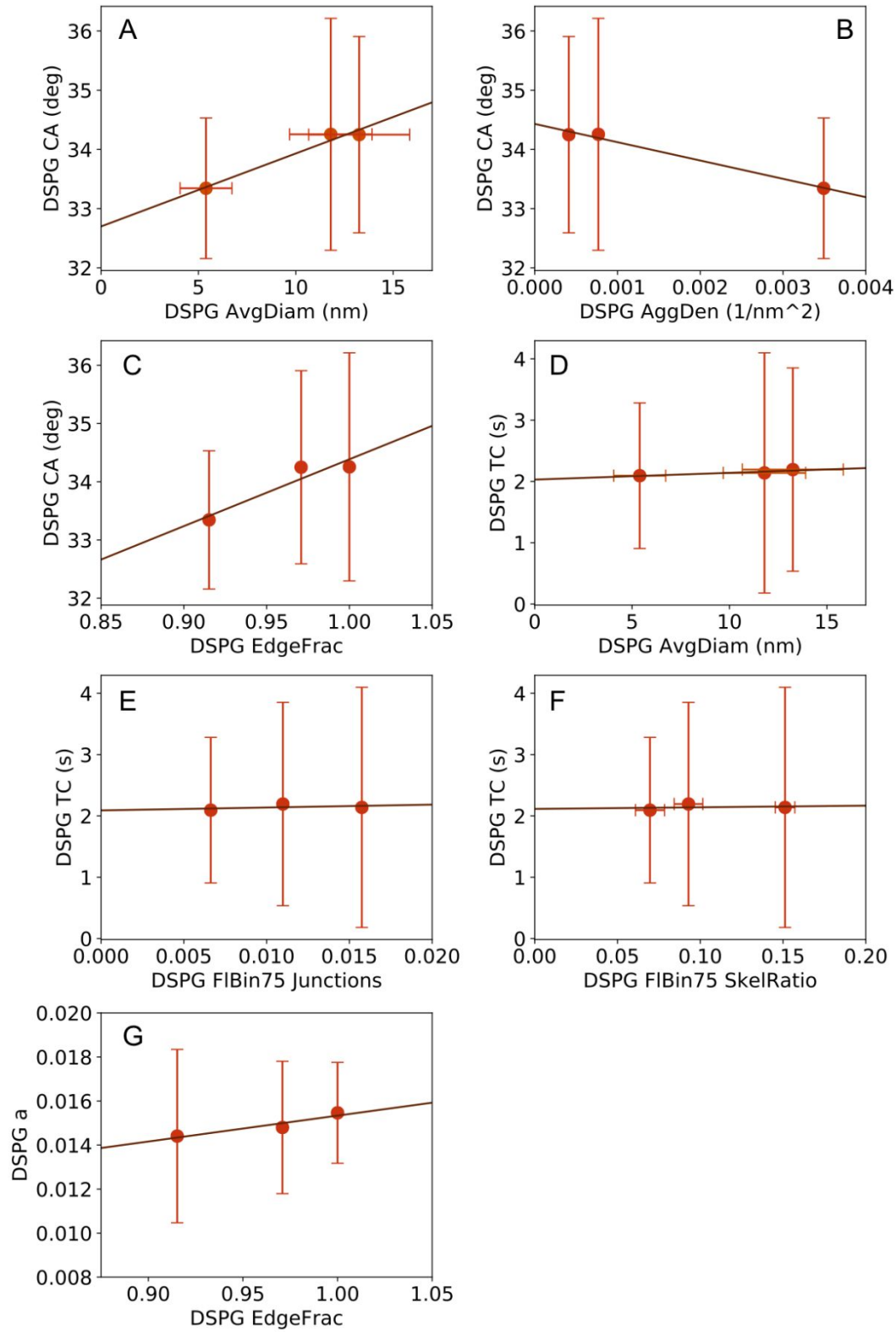


Figure S8: DSPG correlation plots for which $r^2 > 0.8$. Note large error bars and flat a values. (A) DSPG aggregate size vs. CA. (B) DSPG aggregate density vs. CA. (C) DSPG DSPG aggregate edge fraction vs. CA. (D) DSPG aggregate size vs. TC. (E) DSPG junction density (fluorescence) vs. TC. Junction density is

reported as the number of junction pixels per square micron. (F) DSPG skeleton ratio (fluorescence) vs. TC. (G) DSPG aggregate edge fraction vs. a.

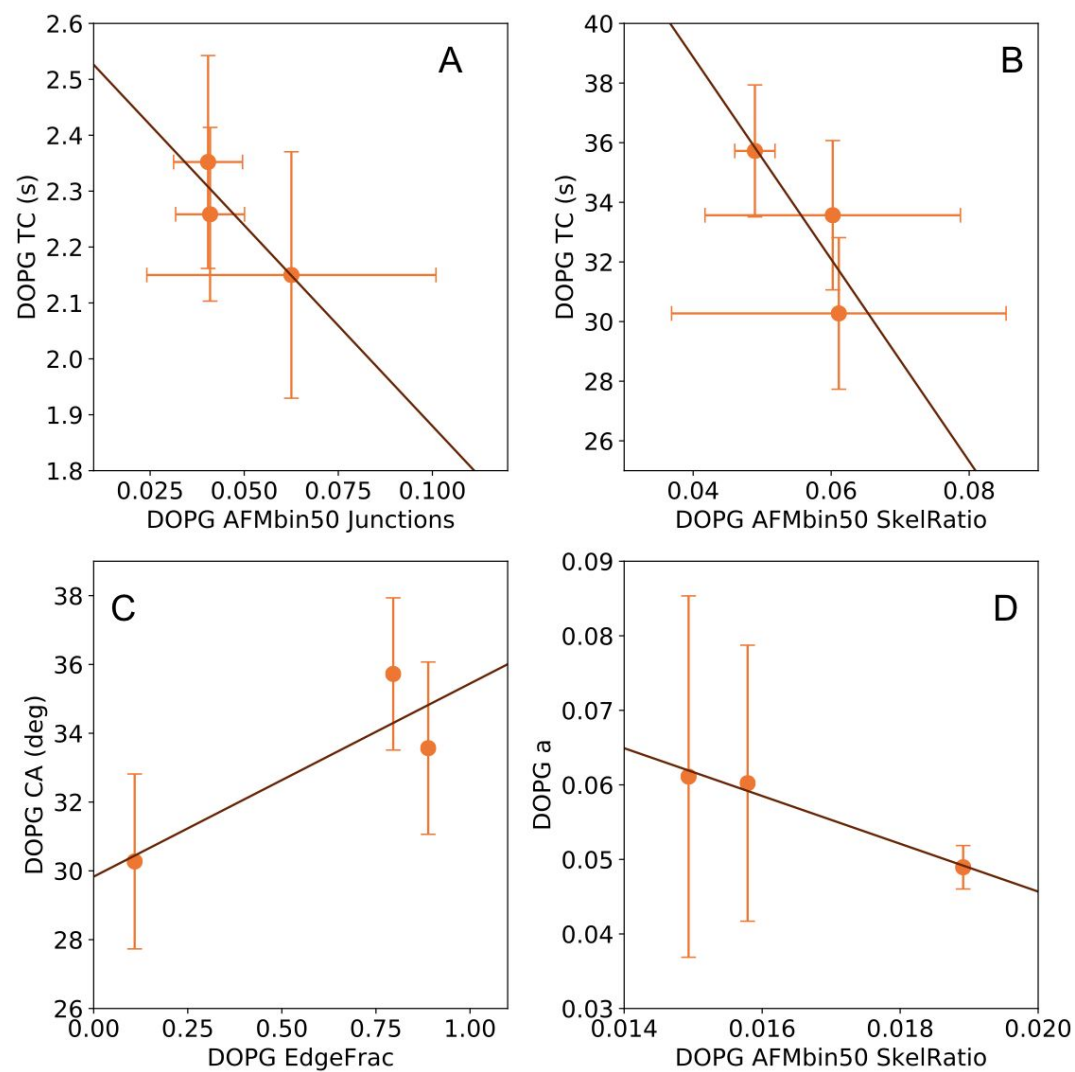


Figure S9: All DOPG correlation plots for which $r^2 > 0.7$. (A) DOPG junction density, fluorescence, vs CA. (B) DOPG skeleton ratio, fluorescence, vs TC. (C) DOPG fraction of aggregates on edges vs CA. (D) DOPG skeleton ratio, AFM, vs. a.