

1 Supplementary Information

1.1 Monolayer

1.1.1 AFM

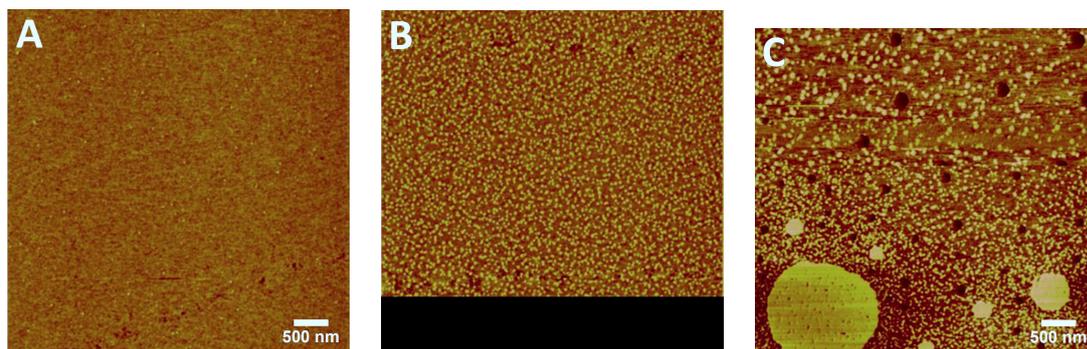


Figure 1: AFM friction images for 20 mol% TASC/DPPC mixture, A. $\Pi = 5$ mN/m, B. $\Pi = 15$ mN/m, C. $\Pi = 35$ mN/m.

1.1.2 Neutron scattering

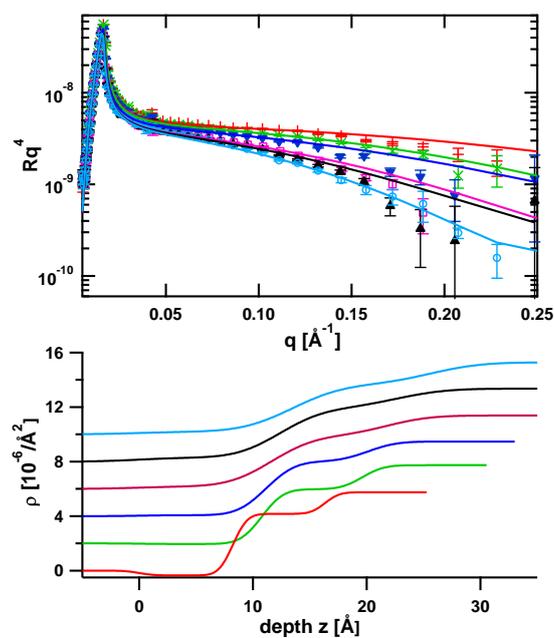


Figure 2: Neutron reflectivity curves and SLD profiles from bottom to top (they are shifted by in steps of 2 to account for better legibility) with increasing surface pressures for TASC/DPPC 2:8 (+ 5 mN/m, × 15 mN/m, ▼ 22 mN/m, □ 30 mN/m, ▲ 35 mN/m, ○ 40 mN/m)

1.2 Bilayer

1.2.1 AFM

1.2.2 Neutron scattering

sample	thickness [Å]	SLD [10^{-6} \AA^{-2}]	roughness [Å]
10 mol% TASC	10.5 ± 1	3.47	5 ± 1
20 mol% TASC	10.5 ± 1	3.47	5 ± 1
50 mol% TASC	9.0 ± 1	3.47	6 ± 1

Table 1: Fitting parameters obtained for SiO₂ layer

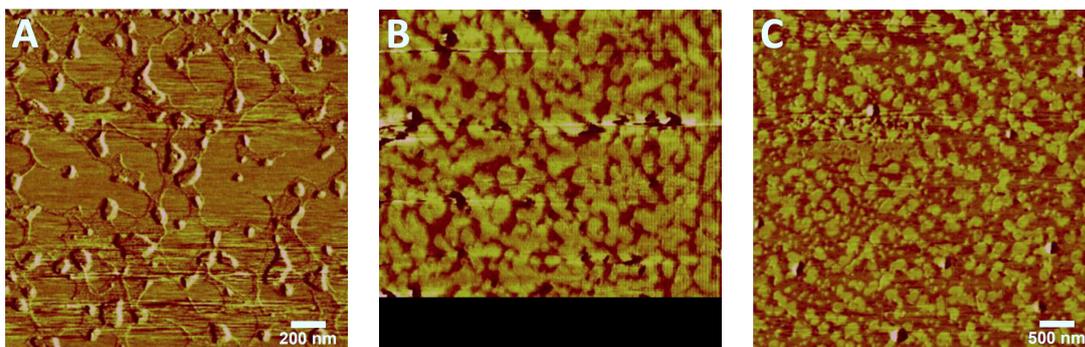


Figure 3: AFM friction images for TASC/DPPC bilayers deposited at 40 mN/m, A. symmetric 10 mol% TASC, B. asymmetric 20 mol% TASC, C. asymmetric 50 mol% TASC.

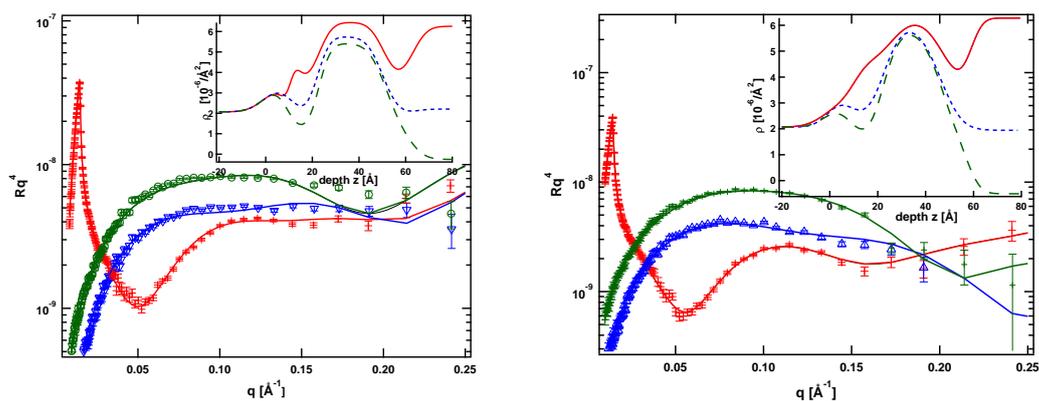


Figure 4: Neutron reflectivity curves for symmetric bilayer with 10 mol% TASC at 25 °C (left) and 55 °C (right) recorded at three contrasts D₂O + H₂O \circ and SMW ∇ , as well as corresponding SDL profiles for D₂O —, H₂O - - - and SMW - - -

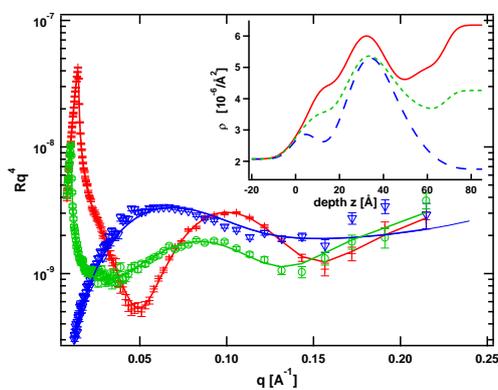


Figure 5: Neutron reflectivity curves for symmetric bilayer with 50 mol% TASC at 25 °C (left) recorded at three contrasts D₂O + 4MW \circ and SMW ∇ , as well as corresponding SDL profiles for D₂O —, 4MW - - - and SMW - - -

symmetric bilayer 10 mol% TASC						
T	layer	water	heads 1	chains 1	heads 2	errors
25 °C	thickness [Å]	1.6	10.0	29.0	12.0	± 1
	SLD [10^{-6}Å^{-2}]	-	1.75	6.44	1.75	± 0.2
	water [%]	100	29	16	39	± 5
	roughness [Å]	6.0	4.8	4.3	5.6	± 1
55 °C	thickness [Å]	1.9	9.5	25.0	11.0	± 1
	SLD [10^{-6}Å^{-2}]	-	1.75	6.52	1.75	± 0.2
	water [%]	100	30	12	37	± 5
	roughness [Å]	5.9	6.3	7.1	7.3	± 1

Table 2: Fitting results for symmetric bilayer 10 mol% TASC

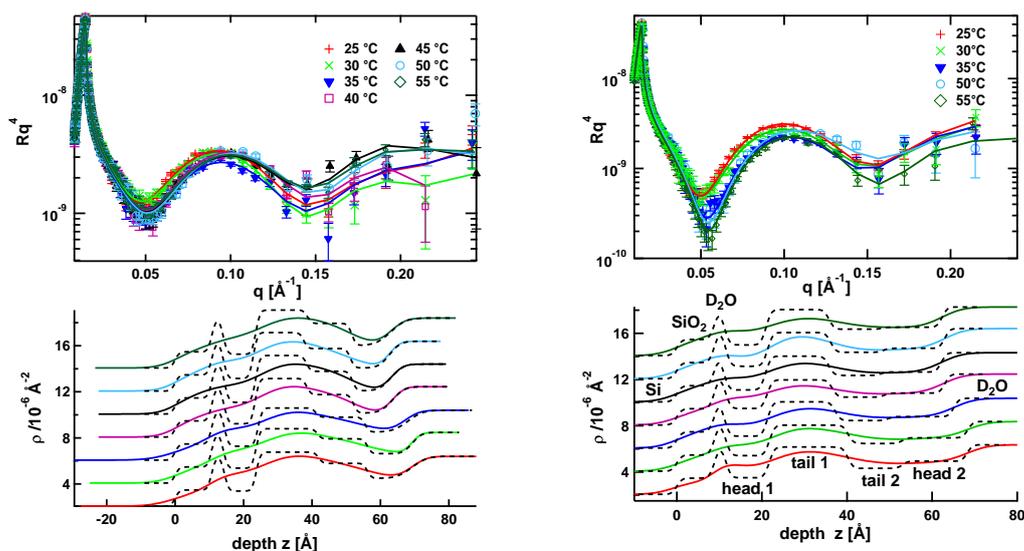


Figure 6: Neutron reflectivity curves for asymmetric bilayers with 20 mol% (left) and 50 mol% TASC (right), as well as corresponding SLD profiles from bottom (25°C) to top (55°C) with increasing temperatures in steps of 5°C. The dashed lines in the SLD profile indicate the different slabs of the box model.

asymmetric bilayer 20 mol% TASC							
T	layer	water	heads 1	chains 1	chains 2	heads 2	errors
25 °C	thickness [Å]	4.0	9.0	18.0	14.0	13.0	±1
	SLD [10^{-6}Å^{-2}]	-	1.72	7.00	5.90	1.83	±0.2
	water [%]	100	36	15	15	58	±5
	roughness [Å]	4.7	7.6	7.5	6.7	7	±1
30 °C	thickness [Å]	4.0	9.5	18.0	14.0	12.5	±1
	SLD [10^{-6}Å^{-2}]	-	1.72	6.85	5.84	1.84	±0.2
	water [%]	100	37	15	15	59	±5
	roughness [Å]	5	7.5	8	5.4	6.7	±1
35 °C	thickness [Å]	3.5	8.5	17.0	14.5	12.5	±1
	SLD [10^{-6}Å^{-2}]	-	1.70	6.46	5.77	1.81	±0.2
	water [%]	100	33	15	15	59	±5
	roughness [Å]	4.6	8.1	8.2	5.6	7.9	±1
40 °C	thickness [Å]	3.5	9.0	15.0	14.0	11.5	±1
	SLD [10^{-6}Å^{-2}]	-	1.68	7.26	5.70	1.81	±0.2
	water [%]	100	28	10	10	48	±5
	roughness [Å]	4.4	6.9	8.2	7.7	6	±1
45 °C	thickness [Å]	3.5	9.0	15.0	14.0	11.5	±1
	SLD [10^{-6}Å^{-2}]	-	1.68	7.30	5.69	1.82	±0.2
	water [%]	100	30	10	10	50	±5
	roughness [Å]	4.5	6.8	8.2	7.9	6	±1
50 °C	thickness [Å]	3.5	8.5	14.5	13.5	11.5	±1
	SLD [10^{-6}Å^{-2}]	-	1.68	7.26	5.70	1.82	±0.2
	water [%]	100	30	10	10	50	±5
	roughness [Å]	4.5	5.4	8.2	7.4	7.1	±1
55 °C	thickness [Å]	3.5	9.5	15.0	13.0	11.5	±1
	SLD [10^{-6}Å^{-2}]	-	1.70	7.18	5.89	1.83	±0.2
	water [%]	100	28	10	10	51	±5
	roughness [Å]	6.0	6.9	8.4	7.7	6.1	±1

Table 3: Fitting results for asymmetric bilayer 20 mol% TASC

asymmetric bilayer 50 mol% TASC							
T	layer	water	heads 1	chains 1	chains 2	heads 2	errors
25 °C	thickness [Å]	3.5	10.0	18.0	13.0	15.0	±1
	SLD [10^{-6} \AA^{-2}]	-	1.72	6.75	3.96	2.02	±0.2
	water [%]	100	31	18	18	68	±5
	roughness [Å]	6.0	6.6	7.8	7.3	8.0	±1
30 °C	thickness [Å]	3.0	10.0	17.5	12.0	15.5	±1
	SLD [10^{-6} \AA^{-2}]	-	1.7	6.63	3.76	2.07	±0.2
	water [%]	100	33	19	19	68	±5
	roughness [Å]	5.8	6.5	8	8	8	±1
35 °C	thickness [Å]	3.5	10.5	15.5	12.0	13.5	±1
	SLD [10^{-6} \AA^{-2}]	-	1.68	6.5	3.72	2.01	±0.2
	water [%]	100	28	2 15	15	62	±5
	roughness [Å]	6	5	8.9	8	7.6	±1
40 °C	thickness [Å]	3.0	10.0	14.5	11.5	11.5	±1
	SLD [10^{-6} \AA^{-2}]	-	1.75	6.55	3.94	1.81	±0.2
	water [%]	100	33	13	13	48	±5
	roughness [Å]	5.2	5.7	8.5	8.1	6	±1
45 °C	thickness [Å]	2.5	10.5	14.0	11.0	13.5	±1
	SLD [10^{-6} \AA^{-2}]	-	1.74	6.52	3.89	2.02	±0.2
	water [%]	100	29	13	31	61	±5
	roughness [Å]	4.9	5.35	9	7.1	7	±1
50 °C	thickness [Å]	3.0	10.0	14.5	11.5	14.0	±1
	SLD [10^{-6} \AA^{-2}]	-	1.68	6.48	3.84	1.96	±0.2
	water [%]	100	29	13	13	61	±5
	roughness [Å]	4.4	5.4	6.4	7	8	±1
55 °C	thickness [Å]	3.0	10.0	14.5	12.0	13.5	±1
	SLD [10^{-6} \AA^{-2}]	-	1.68	6.72	3.93	1.96	±0.2
	water [%]	100	29	12	12	60	±5
	roughness [Å]	5.4	5.8	8.0	8.0	8.33	±1

Table 4: Fitting results for asymmetric bilayer 50 mol% TASC