Electronic Supplementary Material (ESI) for Soft Matter

Formation of supported lipid bilayers on silica: relation to lipid phase transition temperature and liposome size

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Supporting Information

Liposome characterization

In Figure S1, the liposome size distributions as obtained by NTA are shown. The liposomes were prepared by extrusion through filters with different pore sizes, see main text.

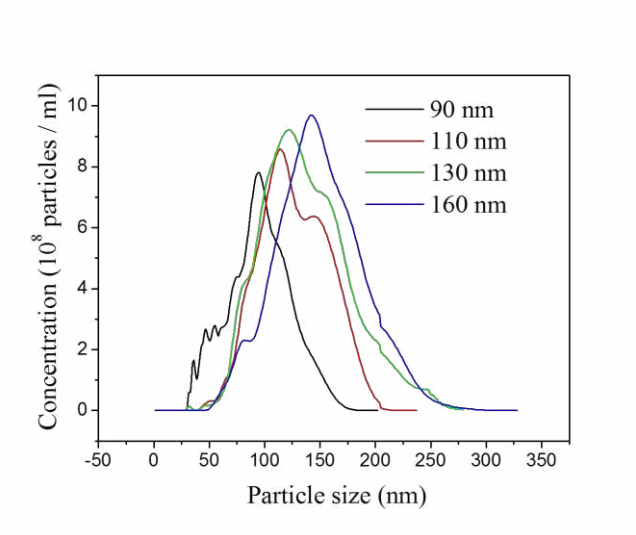

Figure S1. Size distributions of DPPC liposomes (average diameters: 90 nm, 110 nm, 130 nm, 160 nm) recorded by NTA.

Reference subtraction of QCM-D data recorded during temperature-ramping.

Since the QCM-D signals are temperature-dependent, it is important to control the temperature carefully while running the experiments. To properly evaluate experiments where the temperature was varied, it is important to subtract a background reference measurement using the same crystal. The data shown in Figure S2 are the recorded QCM-D frequency and dissipation shifts, while adsorbing DPPC liposomes to a silica surface, and the corresponding measurements using the same crystal without addition of liposomes. The corresponding subtracted data are shown in Figure 4 in the main text.

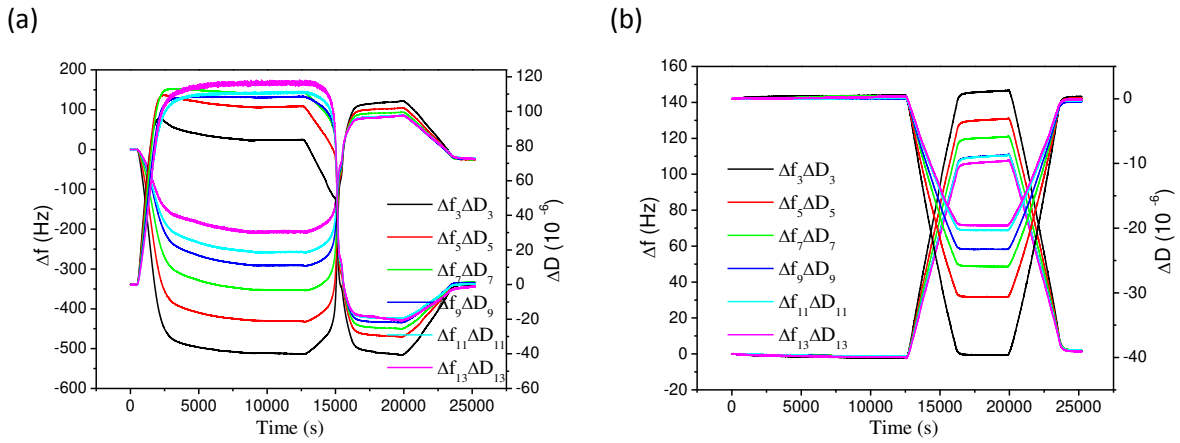

Figure S2. (a) QCM-D frequency and dissipation shifts recorded for 160 nm DPPC liposome (33 % NaCl containing) adsorbing to a silica surface and while subsequently exposed to a precisely controlled temperature profile; (b) Reference data collected using the same crystal under bulk flow and the same temperature condition.

Repeated temperature cycles for liposomes attached at low coverage

Upon repeated tempertaures cycles (Figure S3), the adsorption step and the first temperature cycle showed an almost indentical profile as in Figure 4. When the temperature cycling treatment continued, Δf and ΔD were barely changed at temperatures above 41 °C, but at 22 °C Δf increased and ΔD decreased slightly after each cycle. Figure S4 (b) magnified Δf and ΔD in a single cycle, the schematic illustration depicted a small portion of the liposomes ruptured and form discrete pieces of lipid bilayer membranes.

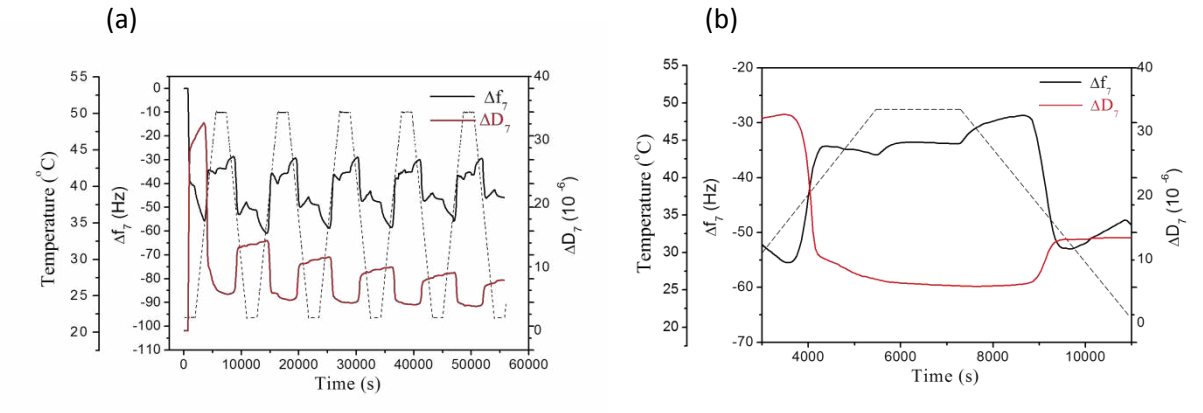

Figure S3. (a) Reference subtracted frequency and dissipation shifts recorded for DPPC liposomes (containing 33% NaCl) partially covered sensor surface under five repeated temperature cycling treatment. (b) Liposome responses in single temperature cycling (temperature profile indicated by dash lines). The data shown were recorded at the 7th overtone.

Viscoelastic modeling of the QCM-D data

In QCM-D measurements, non-rigid films typically display high ΔD/Δf ratios. The layers of intact liposomes in this study clearly belong to this non-rigid regime, especially for the larger liposomes. Furthermore, at low liposome coverage on the sensor surface, the layer is a film of discrete liposomes, which makes the interpretation of ΔD and Δf more difficult. For liposomes undergoing phase transitions, adsorbed liposome layers have previously been successfully represented by a Voigt-based viscoelastic model.[1] Similarly to the previous modeling, we will assume that liposome deformation (and the resulting volume changes) and corresponding membrane states (the membrane mechanical properties will be very different below and above T_m) both influence the QCM-D signals.

Modeling of the QCM-D results using a Voigt-based viscoelastic model readily produce the expected mass differences between similarly sized 33% NaCl-filled and PBS-filled liposomes (Table S1). The following values for densities were used; DPPC lipid 1.05 g/cm^3, PBS buffer 1.00 g/cm^3, 0.56 M NaCl solution 1.30 g/cm^3. By assuming that all the liposomes kept a spherical shape (diameter: 160 nm), the effective density for 33% NaCl-containing DPPC liposomes was estimated to 1.25 g/cm^3, and for PBS-containg DPPC liposomes to 1.00 g/cm^3. The modeled values are relatively close to theoretical estimations, and the fitted effective parameters are consistent with a liposome layer below the

phase transition temperature. The liposomes under less osmotic pressure formed a softer layer compared to the 33% NaCl-filled liposomes.

Table S1. Estimated mass of liposome layers obtained by modeling of the QCM-D results obtained for liposomes adsorbed to silica at 22 ^{o}C. The average liposome size was 160 nm.

	33% NaCl-containing DPPC liposome	PBS-containing DPPC liposome
QCM-D frequency shift[a]; Δf_3 (Hz)	500 ± 10	240 ± 10
QCM-D dissipation shift[a]; ΔD_3 ($\times 10^{-6}$)	29 ± 1	29 ± 1
Mass estimated from Voigt-based modeling[b] (ng/cm^2)	18820 ± 7	9580 ± 14
Theoretical mass[c] (ng/cm^2)	13800	10666
χ^2	39 ± 9	52 ± 18
Effective density (g/cm^3)	1.25	1.00
Effective shear viscosity (g/ms)	3.2	2.17
Effective shear elastic modulus (kPa)	7.0	5.7

[a)] Note that frequency and dissipation shifts are different for different overtones (see Figure S2). Average values and standard deviations were calculated from 5 experiments. [b)]The modeling was based on one experiment with a Δf_3 close to the average value. Averages and standard deviations are presented based on a series of fittings using different starting values for the fitted parameters. [c)] The theoretical mass was calculated assuming that both PBS- and 33% NaCl-containing liposomes were densely packed spheres (diameter: 160 nm).

References

1 G. Ohlsson, A. Tigerström, F. Höök and B. Kasemo, *Soft Matter*, 2011, **7**, 10749-10755.