## **Electronic Supplementary Information**

## Enhancing the Stability of Spontaneously Self-Assembled Vesicles - The Effect of Polymer Architecture

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## Experimental

Kinetic Dynamic Light Scattering Measurements (DLS)



Figure S1: Intensity autocorrelation function  $g^{(2)}$  determined by DLS measurements on the system TDMAO -LiPFOS (55:45, 50 mM) after mixing in the stopped-flow device a) 4 s and b) 1144 s after preparation. Vesicles 4 s after mixing are monodisperse and the intensity autocorrelation function  $g^{(2)}$  can be described by a monoexponential function. The vesicle radius is 8 nm. 1144 s after preparation the vesicle radius increases to 13 nm and vesicle ageing can be observed indicated by a shoulder in the intensity autocorrelation function, which can be described by a double exponential function.



Figure S2: Ratio between the hydrodynamic radius  $R_{\rm h}$  of a vesicle with free diffusion and the apparent hydrodynamic radius  $R_{\rm h,a}$  assuming a hard-sphere potential

## Results

The volume fraction of amphiphilic material  $\phi$  was calculated from the total concentration of amphiphilic material  $c_{\text{tot}}$ , the molar mass  $M_i$  of each component i, and the density  $\rho_i$ .

$$\phi = c_{\rm tot} \cdot \sum_{\rm i} \frac{M_{\rm i}}{\rho_{\rm i}} \tag{S1}$$



Figure S3: Time dependent development of the z-average hydrodynamic radius  $R_{\rm h}$  (mixture: TDMAO:LiPFOS (55:45) 50 mM + Pluronic 10R5, 25 °C)



Figure S4: Hydrodynamic radius at 100 s after mixing as a function of the polymer content (mixture: TDMAO:LiPFOS (55:45) 50 mM + polymer , 25 °C)



Figure S5: Hydrodynamic radius 100 s after mixing (mixture: TDMAO:LiPFOS (55:45) 50 mM + polymer , 25 °C)



Figure S6: Time dependent development of the Rayleigh ratio  $R_{\theta}$  and the vesicle radius  $R_{\text{ves}}$  determined from static light scattering  $R_{\text{ves}}$  comparing Pluronic L35 and Pluronic 10R5 at two different polymer concentrations (mixture: TDMAO:LiPFOS (55:45) 50 mM + polymer, 25 °C); black squares: mixture without polymer, red symbols: mixture with Pluronic L35, green symbols mixture: with Pluronic 10R5, diamonds: c(polymer)=0.01375 mM, circles: c(polymer)=0.1375 mM, triangles: c(polymer)=0.55 mM



Figure S7: Time dependent development of the Rayleigh ratio  $R_{\theta}$  and the vesicle radius  $R_{\text{ves}}$  determined from static light scattering  $R_{\text{ves}}$  (mixture: TDMAO:LiPFOS (55:45) 50 mM + Pluronic F38, 25 °C)



Figure S8: Time dependent development of the Rayleigh ratio  $R_{\theta}$  and the vesicle radius  $R_{\text{ves}}$  determined from static light scattering  $R_{\text{ves}}$ ; mixtures: TDMAO:LiPFOS (55:45) 50 mM: black circles; TDMAO:LiPFOS (55:45) 50 mM + Pluronic F88 (0.0137 mM, 0.0275 mM, 2.75 mM), 25 °C, red crosses; TDMAO:LiPFOS (55:45) 50 mM + Pluronic F108 (0.0137 mM, 0.055 mM, 0.275 mM), 25 °C, green diamonds