Supporting Information: **Shrink-wrap Vesicles**

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**Figure S1.**

Standard curve of donor fluorescence vs. probe density for the FRET assay ($F/F_0 = \text{donor fluorescence/ donor fluorescence in 1\% Triton X-100}$). The solid line shows a fitted curve ($y = -0.203*ln(x) - 0.025$).

**Figure S2.**

Standard curve of self-quenching of calcein vs. concentration ($F_{ves}/F_{ves}^0 = \text{calcein fluorescence/ calcein fluorescence in 1\% Triton X-100}$). The solid line shows a fitted curve ($y = 0.00052*x^2 - 0.036*x + 1.12$).
Release of calcein during and after shrinking of MA vesicles (a representative trial is shown). Although vesicle size has equilibrated by ~500 minutes (see Fig. 1A), calcein continues to be released slowly. The solid line shows a single exponential decay curve fit for the short time course ($k \sim 0.03 \text{ min}^{-1}$).

Final vesicle diameter after shrinking using micelles composed of varying mole percentages of POPC and MA, measured by DLS (~40 mM vesicles + 40 mM micelles).
The fluorescence of calcein encapsulated in MA vesicles after dilution into an isotonic solution (0.2 M bicine, pH 8.5; brown line) or hypertonic solution (0.5 M bicine, pH 8.5; red line). In hypertonic solution, vesicles shrink osmotically, causing the internal concentration of calcein to increase and therefore the fluorescence to become self-quenched.\textsuperscript{1,2} The recovery of fluorescence within several hours indicates the return to a spherical volume, due to equilibration of bicine across the vesicle membrane.

Shrinking of oleate vesicles (~3 mM) after adding MA/POPC (89% MA, 11% POPC) micelles (4 mM).
Supplementary Text

During vesicle growth, MA micelles that are not closely associated with a membrane (i.e., excess beyond 0.4 equivalents) aggregate into large metastable structures. However, during vesicle shrinking, because MA/POPC micelles do not aggregate at pH 8.5, excess MA/POPC micelles would increase the effective ratio of micelles to vesicles. Transfer of MA from vesicles to micelles may then occur in these complexes, a first-order process that would give rise to the observed exponential decay during shrinking.

Supplementary References