

# Transition from complete to partial wetting within membrane compartments

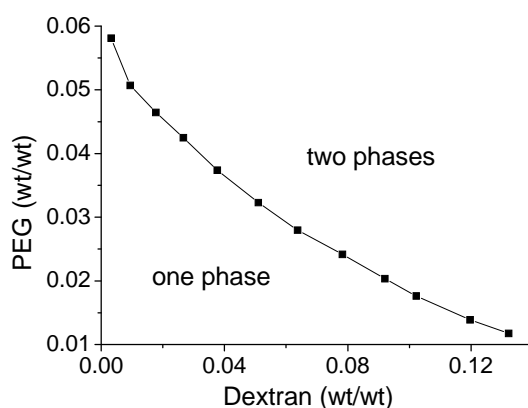
## Supporting Information

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**Materials.** Poly(ethyleneglycol) or PEG (average molecular weight 8000 g/mol) and dextran from Leuconostoc mesenteroides (molecular weight between 400 kDa and 500 kDa) were purchased from Sigma-Aldrich. The polydispersity, measured with gel permeation chromatography, was 1.11 for PEG and 1.83 for dextran. Fluorescein isothiocyanate-dextran (average molecular weight 500,000) was purchased from Sigma. Sucrose was purchased from Fluka. Lipids including 1,2-dioleoyl-sn-glycero-3-phosphocholine (DOPC), and 1,2-dipalmitoyl-sn-glycero-3-phosphoethanolamine-N-(lissamine rhodamine B sulfonyl) (DPPE-Rhod) as chloroform solutions and galbeta1-3galnacbeta1-4(neuacalpha2-3)galbeta1-4glcbeta1-1'-cer ( $G_{M1}$  Ganglioside) as powder were purchased from Avanti Polar Lipids.

**Phase diagram of the polymer aqueous solution in bulk.** The binodal of the PEG and dextran aqueous solution at room temperature was determined by cloud-point titration for each polymer concentration combination. At first, concentrated PEG and dextran aqueous stock solutions were prepared. The mass of a small well sealed vial with a stirring bar inside was measured with a Mettler AT261 DeltaRange balance. Then, a certain amount of dextran stock solution and water was injected into the vial through a syringe, and the added masses were measured. PEG stock solution was injected drop wise into the vial through a syringe under stirring until the solution in the vial became turbid and the added mass was measured again. The same procedure was repeated for several different initial dextran concentrations. The cloud point titration was done at 23°C. One should note that the molecular weight and the polydispersity of the polymers can influence the binodal. In a similar fashion, the vesicle behavior is very sensitive to the concentration and composition of the polymer solutions. In particular, its morphology at these polymer concentrations is different in the absence of labeled dextran.



**Figure S1** Phase diagram of PEG/dextran aqueous solutions at 23°C. The line is a guide to the eyes.

### Giant vesicle preparation using electroformation

25 $\mu$ l stock lipid solution in chloroform (2 mg/ml) was spread on glass substrates coated with indium tin oxide (ITO). The lipid films were dried in a vacuum desiccator for at least 3 hours. A rectangular Teflon frame of thickness 1.6 mm

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served as a spacer between two opposing glass substrates. The chamber was sealed with grease. The coated ITO surfaces acted as electrodes. Circa 2 ml preheated polymer solution was injected into the chamber through a 0.22  $\mu\text{m}$  filter. The chamber was placed in an oven at 60°C and immediately afterwards an AC field of 1.5V (peak-to-peak) and 10 Hz was applied using a function generator (Agilent 33220A 20MHz function/arbitrary waveform generator). The electroformation continued for 2 or 3 hours. The chamber was taken out of the oven and cooled to room temperature (23°C). Then the polymer solution with vesicles was transferred into a small tube. The polymer solution used here (4.05% PEG, 2.22% dextran where 0.52% of total dextran was fluorescein isothiocyanate-dextran) was in one phase at room temperature.

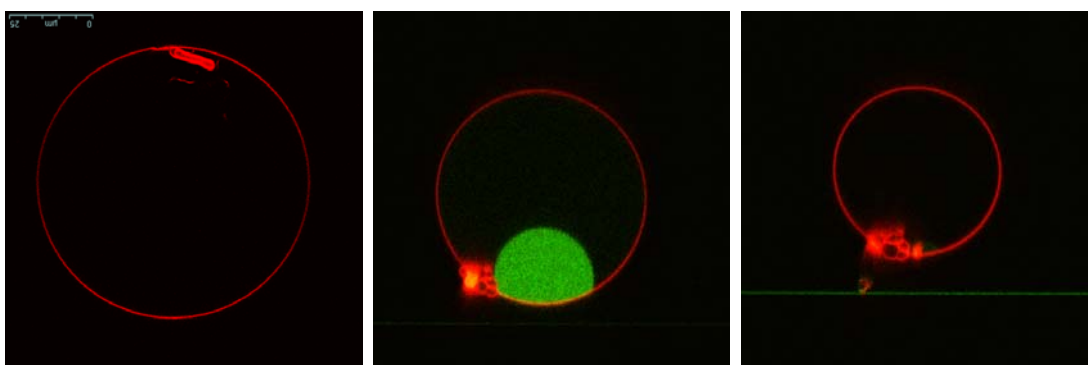
For the calculation of the total polymer concentration in the vesicle we assumed that the initial polymer concentration was the same as in the polymer solution, in which the vesicles were prepared. These two concentrations might differ slightly.<sup>1</sup> The latter will result only in a small shift in the evaluated total polymer concentration but will not affect the wetting transition itself.

### Characterization of vesicles

The vesicles were diluted in the aqueous solution containing 4.41% PEG and 1.45% dextran. This solution had the same osmolarity as the polymer solution used for vesicle formation, but it has a lower density. The osmolarity was measured with an osmometer (Gonotec Osmomat 030).

Phase separation of the polymer solution in vesicles was induced by deflation, namely by injecting a hypertonic aqueous solution into the chamber. The latter was prepared by dissolving 0.1 mole sucrose in 1 liter polymer solution, which corresponds to 3.27% sucrose, 3.92% PEG and 2.14% dextran and an osmolarity of 0.146 Osmol/kg. The deflation was done stepwise.

The behavior of the vesicles after deflating was observed using a confocal microscope (Leica TCS SP5) with a 63 $\times$  water immersion objective at room temperature. The excitation wavelength is 488 nm for the labeled dextran, and 561 nm for the labeled membrane. Due to spherical aberrations, the vesicle images were distorted in the z-direction. This was corrected using correction factors as estimated from scanning cross sections of spherical latex particles in solutions of various refractive indices.



**Figure S2** Vertical cross sections of a vesicle with lipids lumps under different conditions. From left to right, the osmolarity ratio  $r$  is 1, 1.8 and 2.0. There were some defects (tethers and a tube) in the vesicle (the first image) before it was exposed to hypertonic medium. Only the fluorescence signal from the membrane is shown in this image. After the vesicle was exposed to hypertonic medium, the excess area formed a cluster with small vesicles and lipids aggregates (second and third pictures).

(1) Dominak, L. M.; Keating, C. D. *Langmuir* **2007**, *23*, 7148-7154.