Supporting Information to: Morphology Transformations of DODAB Vesicles

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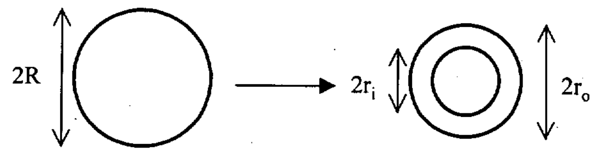
Appendix I

The extent of the size reduction and the degree of displacement of the entrapped volume can be calculated from geometric considerations. When it is assumed that the surface area of the vesicle is fixed, the relevant geometric parameters can be easily obtained:



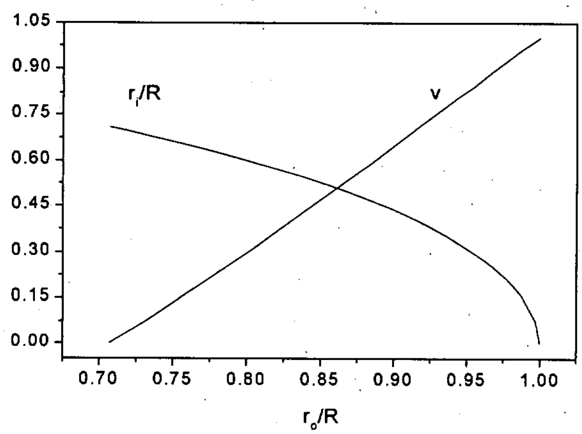
$$4\pi R^2 = 4\pi r_i^2 + 4\pi r_o^2 \qquad [I.1]$$

$$\rho = r_o / R \qquad [I.2]$$

$$r_i / R = \sqrt{1-\rho^2} \qquad [I.3]$$

$$v = \rho^3 - (1-\rho^2)^{3/2} \qquad [I.4]$$

In these definitions R, r_i and r_o stand for the radius of the parental vesicle, and the radius of the inner and outer vesicles of the twinned architecture, respectively. The size reduction following the process of gemination is defined by $\rho = r_o/R$. The ratio of the volume enclosed by the two spherical surfaces and the volume of the parental vesicle is represented by v. In other words, (1-v) represents the fraction of entrapped solution that is forced to permeate through the bilayer upon transformation. It can be recognised that size reduction is limited to $\rho = \sqrt{1/2}$, reflecting the situation where the inner and outer vesicle contact.



S-Figure 1. Geometric considerations on the enclosed volume as a function of the size reduction.

Appendix II

In the Rayleigh-Gans-Debye theory of light scattering, the light intensity scattered per unit volume of solution is[1,2]

$$I(\theta) = \frac{9\pi^2 V^2 N}{r^2}\left(\frac{n}{\lambda_0}\right)^4\left(\frac{m^2-1}{m^2+2}\right)^2(1+\cos^2\theta)P(\theta)I_0 \qquad \text{[II.1]}$$

where V is the volume of the scattering material of a single particle, N is the particle number concentration, r is the observation distance, n is the refractive index of the medium, λ_0 is the wavelength in vacuum of the incident beam, I_0 the incident intensity, $m=n_s/n$ is the ratio of the refractive index of the scattering material (n_s) and of the medium, θ is the angle of observation, and $P(\theta)$ is the scattering factor. The total intensity of the scattered light per unit volume of scattering solution is calculated by integrating formula [II.1] over the surface of a sphere with radius r.

$$S = 24\pi^3 V^2 N \left(\frac{n}{\lambda_0}\right)^4 \left(\frac{m^2-1}{m^2+2}\right)^2 QI_0 = \tau I_0 \quad \text{with} \quad Q = \frac{3}{8}\int_0^{\pi} P(\theta)(1+\cos^2\theta)\sin\theta d\theta \qquad \text{[II.2]}$$

The attenuation of the light due to scattering is simply given by:

$$-dI/dx = \tau I \Rightarrow \ln\left(\frac{I_l}{I_0}\right) = -\tau l \qquad \text{[II.3]}$$

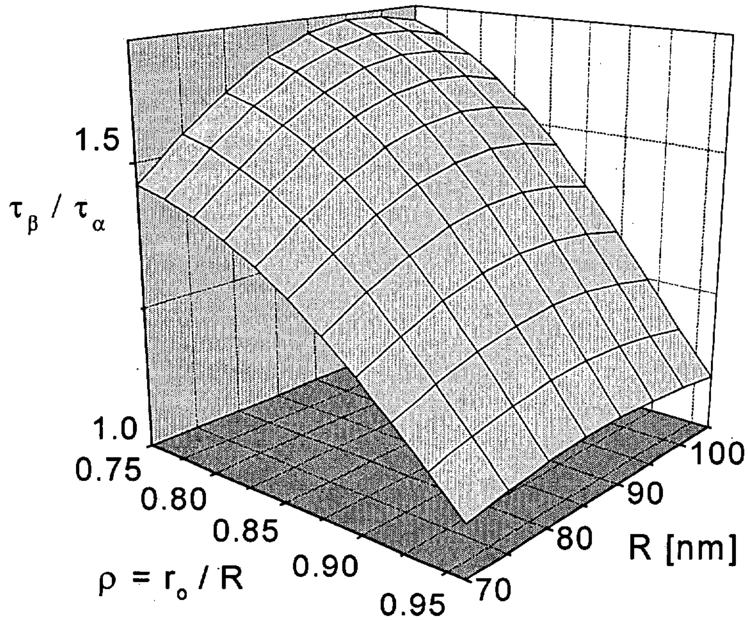
where, I_l is the intensity of the beam emerging at distance l. The attenuation coefficient τ is called the turbidity[2].

The influence of gemination on the turbidity can be calculated. It is noted that during this process no changes in the number concentration (N) and material volume (V) occur. So, the ratio of turbidity is given by the ratio of the dissipation factors: $\tau_\alpha/\tau_\beta=Q_\alpha/Q_\beta$. Subscripts α and β denote the parental and twinned vesicular structures, respectively. Analytical expressions for scattering factors for hollow spherical structures can be found in literature[3,4]. The scattering factor for the vesicle was derived from the formula expression by Oster[5]:

$$P(\theta)_\alpha = \left[\frac{4\pi}{Vk^3}\left(\sin kR - kR\cos kR + k(R-\delta)\cos k(R-\delta) - \sin k(R-\delta)\right)\right]^2 \qquad \text{[II.4]}$$

$$P(\theta)_\beta = \left[\frac{4\pi}{Vk^3}\begin{pmatrix}\sin kr_i - \sin k(r_i-\delta) + k(r_i-\delta)\cos k(r_i-\delta) - kr_i\cos kr_i + \\ \sin kr_o - \sin k(r_o-\delta) + k(r_o-\delta)\cos k(r_o-\delta) - kr_o\cos kr_o\end{pmatrix}\right]^2 \qquad \text{[II.5]}$$

with $k=4\pi n/\lambda_0 \sin(\theta/2)$, referred to as the scattering vector. As before, r_i, r_o and δ stand for the radius of the inner and outer vesicle of the twinned vesicular structure and bilayer thickness, respectively. In conclusion, this set of equations allows the calculation of the influence of gemination as a function of the actual degree of size reduction and the wavelength of incident light.



S-Figure 2. Increase in turbidity as a function of the radius of the parental vesicle R and the degree of size reduction ρ after gemination (r_o stands for the radius of the outer vesicle of the twin) λ=400nm, δ=5nm. τ_α/τ_β represents the ratio of the turbidity of the twinned vesicles and the parental vesicle.

References.

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