# Supporting Information

## 1 Details of Monte Carlo simulations

All the Monte Carlo (MC) simulations are performed by using vesicles with $N_\mathrm{t} = 2000$ triangles (with $N_\mathrm{v} = 1002$ vertices and $N_\mathrm{e} = 3000$ edges) and fixed total area $A_0$. We define the radius $R_0 \equiv \sqrt{A_0/(4\pi)}$ of the reference spherical vesicle which has the same area as the simulated vesicle but different volume $V_0 \equiv 4\pi R_0^3/3$. For simplicity, one can choose the volume $V_0 = 1$ for the reference spherical vesicle. Then the average area $\bar{A}_I$ of all the triangles $I$, defined as

$$\bar{A}_I \equiv \frac{A_0}{N_\mathrm{t}}, \tag{S1}$$

is already known as a constant. By making the area of the triangle shown in Fig. S4 equal to $\bar{A}_I$, one obtains $l_\mathrm{hc} = \sqrt{4\bar{A}_I/\sqrt{3}}$ for the hard-core diameter of the tether potential. In the case with volume constraint, we use the reduced volume $v \equiv V/V_0$. The temperature $T$ is chosen as the basic energy unit. According to the experimental results (see Table 1 in ref. 26), the bending moduli for $\mathcal{L}_d$ and $\mathcal{L}_o$ phases vary with the order of several tens of thermal energy $T$ at experimental temperatures. Without loss of generality, we choose the bending rigidity $\kappa^{(\beta)} = 10\,T$ and vary $\kappa^{(\alpha)}$ by changing the ratio $\kappa^{(\alpha)}/\kappa^{(\beta)}$. Within acceptable cost of computation, all the MC morphologies in the paper are determined from 6–10 MC simulations each having 1.2–2.4$\times 10^7$ MC steps starting from independent random initial configurations. Each MC step consists of $N_\mathrm{v}$ vertex displacements, $3N_\mathrm{e}$ edge flips, and $N_\mathrm{t}$ triangle exchanges.

## 2 Supporting Figures

Figures S1 and S2 present the morphology diagrams as a function of (i) area fraction of the liquid-ordered ($\mathcal{L}_o$) or $\alpha$ phase $\chi^{(\alpha)}$ and (ii) bending rigidity ratio $\kappa^{(\alpha)}/\kappa^{(\beta)}$. Figure S3 presents the energy decomposition of the total energy as obtained by energy minimization in Fig. 3(b). Figure S4 shows the geometry of the triangle which is used to determine the hard-core diameter $l_\mathrm{hc}$ of the tether potential for the triangulated vesicle.

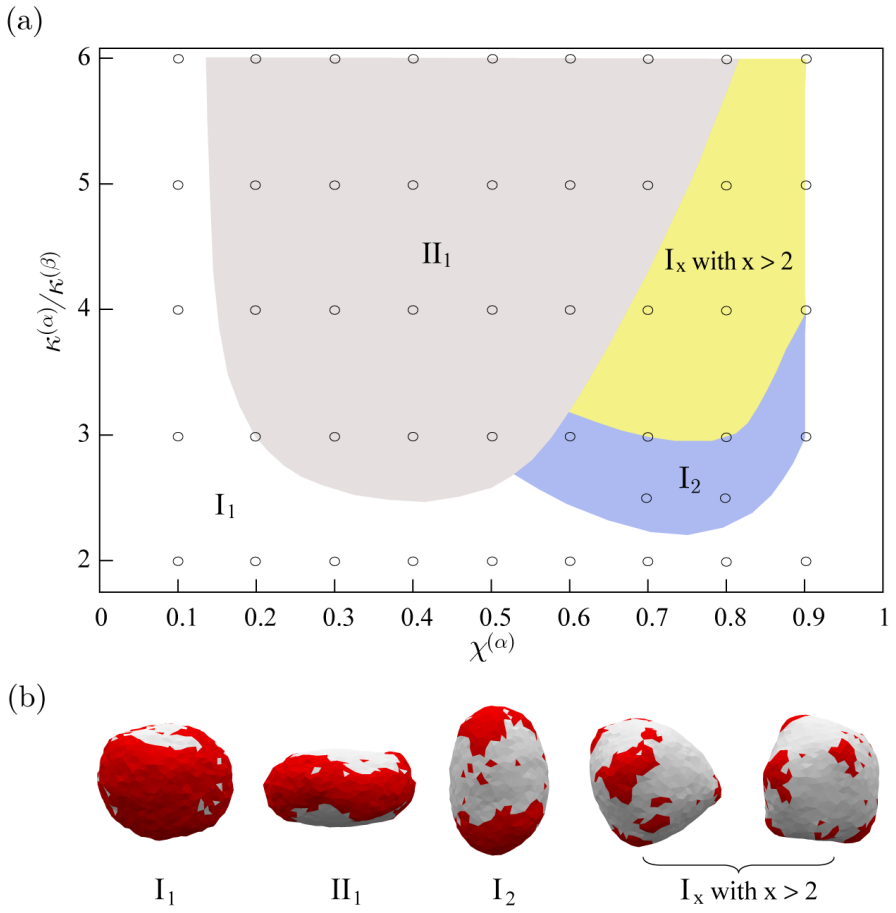


Figure S1: (a) Morphology diagram as a function of (i) area fraction of the $\mathcal{L}_o$ or $\alpha$ phase $\chi^{(\alpha)}$ and (ii) bending rigidity ratio $\kappa^{(\alpha)}/\kappa^{(\beta)}$ as obtained from Monte Carlo simulations of vesicles without volume constraint as well as vanishing spontaneous curvature and equal Gaussian moduli. The interaction parameter is $U = 1.2\,U_c$ with the critical value $U_c$ as given by eqn (17). The open circles in the morphology diagrams represent the parameter values for which simulations have been performed. The transition lines between different morphologies shaded in different colors are rough estimates and include those parameter values for which two stable morphologies have been observed. The notation for the morphologies is explained in the text. In the yellow parameter regime, strong compositional fluctuations lead to two or more metastable morphologies for the same vesicle. For simplicity, we denote those morphologies as $I_x$ with $x > 2$ where the vesicle consists of one majority $\mathcal{L}_o$ or $\alpha$ domain and more than two $\mathcal{L}_d$ or $\beta$ domains. (b) Snapshots from the Monte Carlo simulations in (a). The $\mathcal{L}_o$ or $\alpha$ phase is shown in light grey and the $\mathcal{L}_d$ or $\beta$ phase in red.

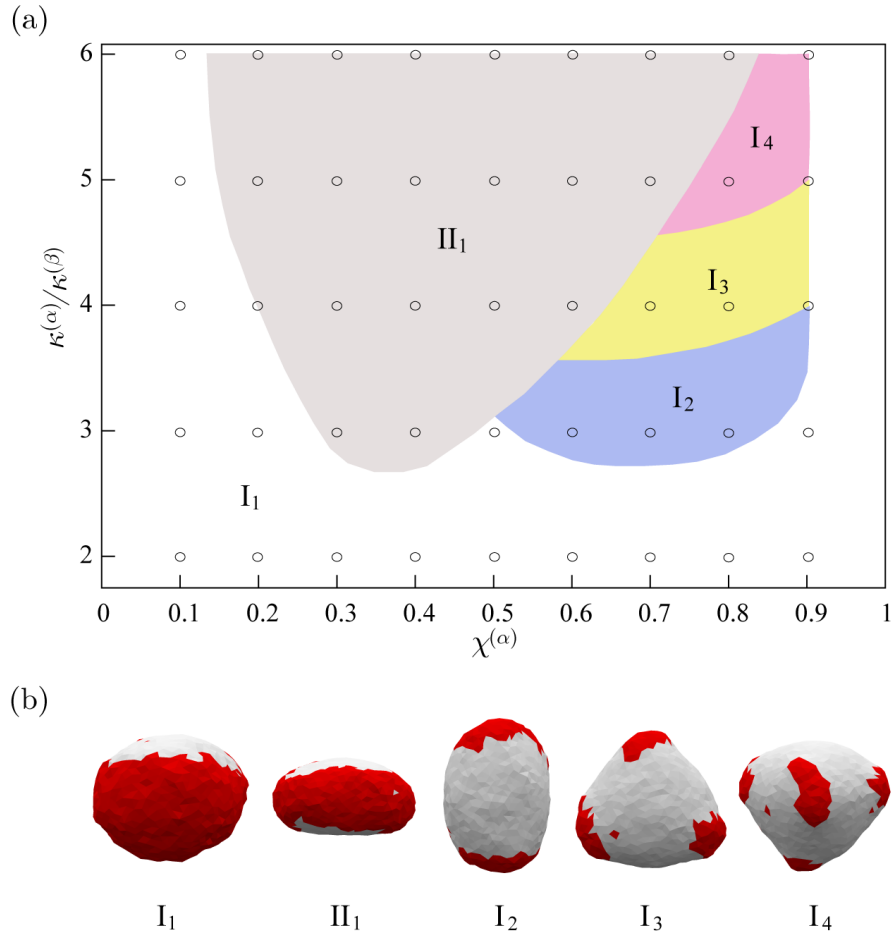


Figure S2: (a) Morphology diagram as a function of (i) area fraction of the $\mathcal{L}_o$ or $\alpha$ phase $\chi^{(\alpha)}$ and (ii) bending rigidity ratio $\kappa^{(\alpha)}/\kappa^{(\beta)}$ as obtained from Monte Carlo simulations of vesicles without volume constraint as well as vanishing spontaneous curvature and equal Gaussian moduli. The interaction parameter is $U = 1.5\,U_c$ with the critical value $U_c$ as given by eqn (17). The open circles in the morphology diagrams represent the parameter values for which simulations have been performed. The transition lines between different morphologies shaded in different colors are rough estimates and include those parameter values for which two stable morphologies have been observed. The notation for the morphologies is explained in the text. (b) Snapshots from the Monte Carlo simulations in (a). The $\mathcal{L}_o$ or $\alpha$ phase is shown in light grey and the $\mathcal{L}_d$ or $\beta$ phase in red. Note that there are less $\alpha$ ($\beta$) triangles embedded in $\beta$ ($\alpha$) domains here than in Fig. S1(b) since the compositional fluctuations are suppressed by increasing interaction parameter $U$ and thus line tension.

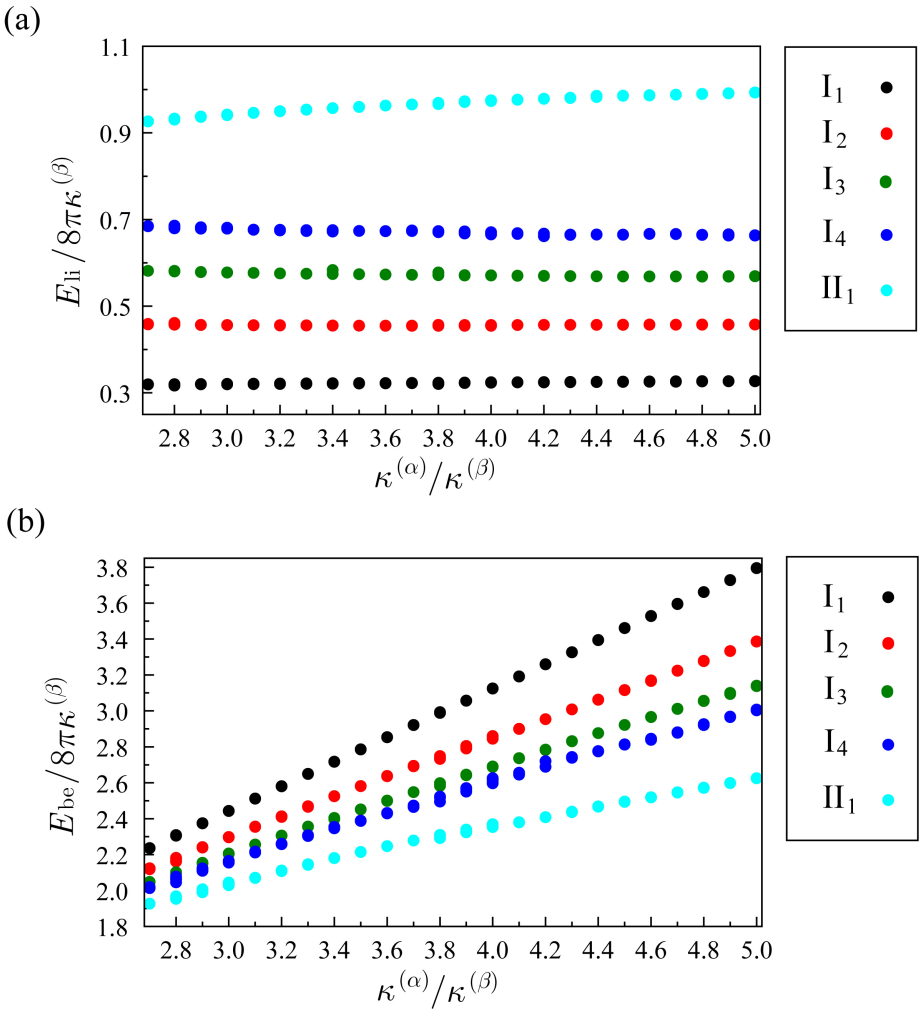


Figure S3: (a) Line tension energy $E_{\rm li}/8\pi\kappa^{(\beta)}$ and (b) bending energy $E_{\rm be}/8\pi\kappa^{(\beta)}$ as a function of bending rigidity ratio $\kappa^{(\alpha)}/\kappa^{(\beta)}$. The sum of these two energies is equal to the total energy $E/8\pi\kappa^{(\beta)}$ as shown in Fig. 3(b).

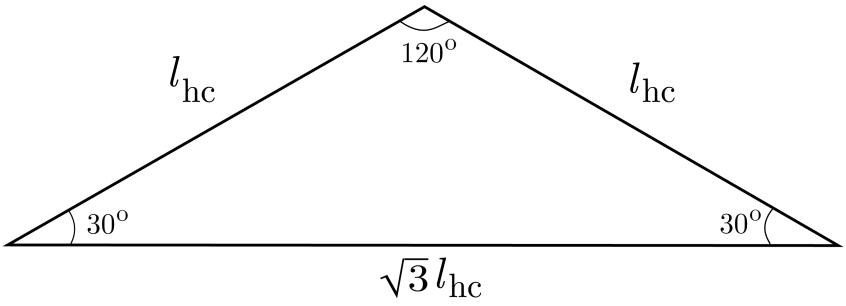


Figure S4: Triangle used to determine the hard-core diameter $l_{\mathrm{hc}}$ of the tether potential for neighbor vertices in the triangulated vesicle.