Figure S1. The variations of (a) pressures and (b) chemical potentials of water in the inner and outer water regions versus simulation steps are shown.
Figure S2. The snapshots of the infinitely large vesicle and its physical properties.

\[
\begin{align*}
    h_\infty &= 5.66 & l_\infty &= 2.92 \\
    \sigma_\infty &= 0.78 & \nu_\infty &= 4.10 \\
    u_\infty &= 12.9 & S_\infty &= 0.62
\end{align*}
\]
Figure S3. The interfacial internal energy per lipid for the inner and outer layers versus the vesicle size ($r$).
Figure S4. (a) The included angle between two neighboring hydrophobic tails; (b) the schematic demonstration of the cylindrical domain for determining the total number of neighboring hydrophobic tails of the centered tail.
Figure S5. $\langle \cos \phi \rangle$ of the inner and outer leaflets are plotted as a function of the vesicle size ($r$). In the inset, $\langle \cos \phi \rangle$ is plotted against $1/r^2$. 