

Supporting Material for "Mimicking effects of cholesterol in lipid bilayer membranes by self-assembled amphiphilic block copolymers"

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1 The model

We calculated the variation of the chemical potential of cholesterol in bilayer membranes, by using a polymeric self-consistent field theory (SCFT) formulated in the canonical ensemble (Tao *et al.* (2007); Cai *et al.* (2017, 2019); Xu *et al.* (2019)). The system is composed of three components: n_{AB} AB diblock (a rod-coil) in which A and B copolymers model a phospholipid (A is the polar headgroup, B is the hydrophobic tail), n_{hA} hA-homopolymer (a coil) that models the external solution (*i.e.*, water), and n_C C-homopolymer (a rod) that models cholesterol. The degree of polymerization is $N = N_A + N_B$, N_{hA} and N_C (see Figure 1). We require that the monomer density ρ_0 is constant to ensure

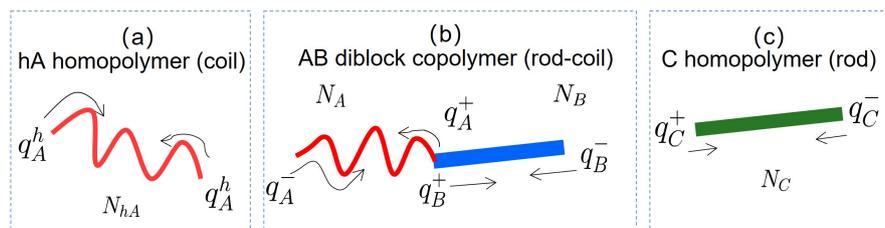


Figure 1: Schematic of the polymers. (a) hA homopolymers with a polymerization degree of N_{hA} . (b) AB diblock copolymers with a polymerization degree of $N = N_A + N_B$. (c) C homopolymers with a polymerization degree of N_C . The arrows indicate the directions that we solve the propagators.

that the bilayer is incompressible. The configurations of the hA homopolymer is represented by the continuous curve $\mathbf{R}_{\alpha^{hA}}(s)(\alpha^{hA} = 0, 1, \dots, n_{hA})$, the C homopolymer is represented by $\mathbf{R}_{\alpha^C}(s)(\alpha^C = 0, 1, \dots, n_C)$, and the AB diblock copolymer is represented by $\mathbf{R}_{\alpha^{AB}}^A(s) = \mathbf{R}_{\alpha^{AB}}^B(s)(\alpha^{AB} = 0, 1, \dots, n_{AB})$, where $s = 0, 1, \dots, N_w$ ($w = hA, AB, C$) is arc length. Gaussian chains are used to model flexible coil polymers; worm-like chains with a rigid parameter λ are used to model rigid rod polymers. The microparticle density distributions and orientational order parameters at position \mathbf{r} is

given

$$\hat{\phi}_A^{hA}(\mathbf{r}) = \frac{1}{\rho_0} \sum_{\alpha_{hA}=1}^{n_{hA}} \int_0^{N_{hA}} ds \delta(\mathbf{r} - \mathbf{R}_{\alpha_{hA}}(s)), \quad (1)$$

$$\hat{\phi}_A^{AB}(\mathbf{r}) = \frac{1}{\rho_0} \sum_{\alpha_{AB}=1}^{n_{AB}} \int_0^{N_A} ds \delta(\mathbf{r} - \mathbf{R}_{\alpha_{AB}}^A(s)), \quad (2)$$

$$\hat{\phi}_A(\mathbf{r}) = \hat{\phi}_A^{hA}(\mathbf{r}) + \hat{\phi}_A^c(\mathbf{r}), \quad (3)$$

$$\hat{\phi}_B(\mathbf{r}) = \frac{1}{\rho_0} \sum_{\alpha_{AB}=1}^{n_{AB}} \int_0^{N_B} ds \delta(\mathbf{r} - \mathbf{R}_{\alpha_{AB}}^B(s)), \quad (4)$$

$$\hat{\phi}_C(\mathbf{r}) = \frac{1}{\rho_0} \sum_{\alpha^C=1}^{n_C} \int_0^{N_C} ds \delta(\mathbf{r} - \mathbf{R}_{\alpha^C}(s)), \quad (5)$$

$$\hat{\mathbf{S}}_B(\mathbf{r}) = \frac{1}{\rho_0} \sum_{\alpha_{AB}=1}^{n_{AB}} \int_0^{N_B} ds \delta(\mathbf{r} - \mathbf{R}_{\alpha_{AB}}^B(s)) \left[\mathbf{u}_{\alpha_{AB}}(s) \mathbf{u}_{\alpha_{AB}}(s) - \frac{\mathbf{I}}{3} \right], \quad (6)$$

$$\hat{\mathbf{S}}_C(\mathbf{r}) = \frac{1}{\rho_0} \sum_{\alpha^C=1}^{n_C} \int_0^{N_C} ds \delta(\mathbf{r} - \mathbf{R}_{\alpha^C}(s)) \left[\mathbf{u}_{\alpha^C}(s) \mathbf{u}_{\alpha^C}(s) - \frac{\mathbf{I}}{3} \right]. \quad (7)$$

Both isotropic and anisotropic interaction are present in the system. The isotropic interaction potential between polymers is given by (Flory (1953); Ohta and Kawasaki (1986); Holyst and Schick (1992))

$$\begin{aligned} H_{isotropic} = & \rho_0 \int_V d\mathbf{r} [e_{AB} \hat{\phi}_A(\mathbf{r}) \hat{\phi}_B(\mathbf{r}) + e_{AC} \hat{\phi}_A(\mathbf{r}) \hat{\phi}_C(\mathbf{r}) \\ & + e_{BC} \hat{\phi}_B(\mathbf{r}) \hat{\phi}_C(\mathbf{r}) + e_{AA} \hat{\phi}_A(\mathbf{r}) \hat{\phi}_A(\mathbf{r}) \\ & + e_{BB} \hat{\phi}_B(\mathbf{r}) \hat{\phi}_B(\mathbf{r}) + e_{CC} \hat{\phi}_C(\mathbf{r}) \hat{\phi}_C(\mathbf{r})], \end{aligned} \quad (8)$$

where $e_{ij} (i, j = \{A, B, C\})$, are the intermolecular interaction forces between i monomers and j monomers, $\chi_{ij} = e_{ij} - (e_{ii} + e_{jj})/2$ is the Flory-Huggins parameter which depends on the polymers and is inversely proportional to temperature (Flory (1953)). The anisotropic potential (Maier-Saupe potential) between molecules reads as (Tao *et al.* (2007); Maier and Saupe (1958); Singh *et al.* (1994); Li *et al.* (2014))

$$H_{anisotropic} = -\frac{\eta \rho_0}{2} \int_V d\mathbf{r} [\hat{\mathbf{S}}(\mathbf{r}) : \hat{\mathbf{S}}(\mathbf{r})], \quad (9)$$

where η is the Maier-Saupe parameter that describes the strength of orientation, and $\hat{\mathbf{S}}(\mathbf{r}) = \hat{\mathbf{S}}_B(\mathbf{r}) + \hat{\mathbf{S}}_C(\mathbf{r})$ is the orientational order parameter. This Maier-Saupe interaction promotes the rod's parallel arrangement. The stretch energy of flexible polymers given by

$$H_{stretch} = \frac{3}{2a^2} \left(\sum_{\alpha_{hA}=1}^{n_{hA}} \int_0^{N_{hA}} ds \left| \frac{d\mathbf{R}_{\alpha_{hA}}(s)}{ds} \right|^2 + \sum_{\alpha_{AB}=1}^{n_{AB}} \int_0^{N_A} ds \left| \frac{d\mathbf{R}_{\alpha_{AB}}^A(s)}{ds} \right|^2 \right), \quad (10)$$

where a is the statistical segment length of hA and A coils. The rigid rods have bending energy that is given by

$$H_{bend} = \frac{\lambda_B}{2b} \sum_{\alpha_{AB}=1}^{n_{AB}} \int_0^{N_B} ds \left| \frac{d\mathbf{u}_{\alpha_{AB}}(s)}{ds} \right|^2 + \frac{\lambda_C}{2c} \sum_{\alpha^C=1}^{n_C} \int_0^{N_C} ds \left| \frac{d\mathbf{u}_{\alpha^C}(s)}{ds} \right|^2, \quad (11)$$

where $\lambda_k (k \in \{B, C\})$ is the persistence length parameter of the wormlike chain used to simulate k rods. b and c are the statistical segment lengths of B and C rods, respectively. $\mathbf{u}_{\alpha_{AB}}(s) = \frac{1}{b} \left| \frac{d\mathbf{R}_{\alpha_{AB}}^B(s)}{ds} \right|$ and $\mathbf{u}_{\alpha^C}(s) = \frac{1}{c} \left| \frac{d\mathbf{R}_{\alpha^C}(s)}{ds} \right|$ are unit orientational vectors. In the canonical ensemble, the system consists of a rod-coil diblock copolymer and a blend homopolymer.

The number of n_{hA} , n_{AB} and n_C is fixed, yielding $V = (n_{hA}N_{hA} + n_{AB}N_{AB} + n_CN_C)/\rho_0$. The partition function for the mixed systems is (Helfand (1975); Edwards (1965))

$$\begin{aligned}
Z = & \frac{1}{n_{AB}!n_{hA}!n_C!} \int D\mathbf{R}_{\alpha^{AB}}^A D\mathbf{R}_{\alpha^{AB}}^B D\mathbf{R}_{\alpha^{hA}} D\mathbf{u}_{\alpha^{AB}} D\mathbf{R}_{\alpha^C} D\mathbf{u}_{\alpha^C} \\
& \times \exp(- (H_{isotropic} + H_{anisotropic} + H_{stretch} + H_{bend})) \\
& \times \delta[\mathbf{u}_{\alpha^{AB}}(s) - \frac{1}{b} \frac{d\mathbf{R}_{\alpha^{AB}}^B}{ds}] \delta[|\mathbf{u}_{\alpha^{AB}}| - 1] \\
& \times \delta[\mathbf{u}_{\alpha^C} - \frac{1}{c} \frac{d\mathbf{R}_{\alpha^C}}{ds}] \delta[|\mathbf{u}_{\alpha^C}| - 1] \\
& \times \delta[\hat{\phi}_A(\mathbf{r}) + \hat{\phi}_B(\mathbf{r}) + \hat{\phi}_C(\mathbf{r}) - 1].
\end{aligned} \tag{12}$$

where $\int D\mathbf{R}$ is an integral over all functions \mathbf{R} , and δ is the Kronecker delta function.

We denote an ensemble average as $\phi_i(\mathbf{r}) = \langle \hat{\phi}_i(\mathbf{r}) \rangle$ of $\hat{\phi}_i(\mathbf{r})$ ($i = A, B, C$), and use the delta function (Fredrickson *et al.* (2006))

$$G(\hat{\phi}_i) = \int D\phi [\delta(\phi_i - \hat{\phi}_i) G(\phi_i)]. \tag{13}$$

to carry out a field transformation

$$\delta(\phi_i(\mathbf{r}) - \hat{\phi}_i(\mathbf{r})) = \int D\omega_i \exp\left(\rho_0 \int_V d\mathbf{r} \omega_i(\mathbf{r}) [\phi_i(\mathbf{r}) - \hat{\phi}_i(\mathbf{r})]\right). \tag{14}$$

We invoke the condition of incompressibility

$$\delta\left(\sum_i \hat{\phi}_i(\mathbf{r}) - 1\right) = \int D\xi \exp\left(\rho_0 \int_V d\mathbf{r} \xi(\mathbf{r}) \left[\sum_i \phi_i(\mathbf{r}) - 1\right]\right). \tag{15}$$

and use the Gaussian integral formulas

$$\exp\left(\frac{\eta\rho_0}{2} \int_V d\mathbf{r} \hat{\mathbf{S}}(\mathbf{r}) : \hat{\mathbf{S}}(\mathbf{r})\right) = \frac{\int D\mathbf{M} \exp\left(-\frac{1}{2\eta\rho_0} \int_V d\mathbf{r} \mathbf{M}(\mathbf{r}) : \mathbf{M}(\mathbf{r}) + \int_V d\mathbf{r} \mathbf{M}(\mathbf{r}) : \hat{\mathbf{S}}(\mathbf{r})\right)}{\int D\mathbf{M} \exp\left(-\frac{1}{2\eta\rho_0} \int_V d\mathbf{r} \mathbf{M}(\mathbf{r}) : \mathbf{M}(\mathbf{r})\right)}. \tag{16}$$

Substituting Eq. (1)-(11) and (13)-(16) into Eq. (12), yields

$$Z = \int \prod_{i \in \{A, B, C\}} D\omega_i D\phi_i D\xi D\mathbf{M} \exp(-H(\omega_i, \phi_i, \xi, \mathbf{M})) \tag{17}$$

where $H(\omega_i, \phi_i, \xi, \mathbf{M})$ is the effective Hamiltonian (the Stirling formula $n! \approx (\frac{n}{e})^n$ (Wu (2016)) is used). This Hamiltonian is defined as

$$\begin{aligned}
H(\omega_i, \phi_i, \xi, \mathbf{M}) = & \rho_0 \int_V d\mathbf{r} \left[\sum_{i \neq j} \chi_{ij} \phi_i(\mathbf{r}) \phi_j(\mathbf{r}) - \sum_i \omega_i \phi_i(\mathbf{r}) + \frac{1}{2\eta} \mathbf{M}(\mathbf{r}) : \mathbf{M}(\mathbf{r}) - \xi(\mathbf{r}) \left(\sum_i \phi_i(\mathbf{r}) - 1\right) \right] \\
& - n_{AB} \log \frac{eV Q_{AB}(\omega_A, \omega_B, \mathbf{M})}{n_{AB}} - n_{hA} \log \frac{eV Q_{hA}(\omega_A)}{n_{hA}} - n_C \log \frac{eV Q_C(\omega_C, \mathbf{M})}{n_C} \\
& + \frac{1}{2} e_{AA} n_{hA} N_{hA} + \frac{1}{2} e_{AA} n_{AB} N_A + \frac{1}{2} e_{BB} n_B N_B + \frac{1}{2} e_{CC} n_C N_C.
\end{aligned} \tag{18}$$

Q_{AB}, Q_{hA}, Q_C are single-chain partition functions corresponding to the AB, hA and C polymer in the fields ω_i, ϕ_i, ξ , and \mathbf{M} . With this notation

$$\begin{aligned}
Q_{AB}(\omega_A, \omega_B, \mathbf{M}) = & \frac{1}{V} \int \prod_{\tilde{k} \in \{A, B\}} D\mathbf{R}^{\tilde{k}} D\mathbf{u}_{\alpha^{AB}} \delta[\mathbf{u}_{\alpha^{AB}} - \frac{1}{b} \frac{d\mathbf{R}_{\alpha^{AB}}^B}{ds}] \delta[|\mathbf{u}_{\alpha^{AB}}| - 1] \\
& \times \exp\left(-\frac{3}{2a^2} \int_0^{N_A} \left|\frac{d\mathbf{R}_{\alpha^{AB}}^A}{ds}\right|^2 ds - \frac{\lambda_B}{2b} \int_0^{N_B} \left|\frac{d\mathbf{u}_{\alpha^{AB}}}{ds}\right|^2 ds\right) \\
& \times \exp\left(-\sum_{\tilde{k} \in \{A, B\}} \int_0^{N_{\tilde{k}}} \omega_{\tilde{k}} ds + \int_0^{N_B} \mathbf{M} : [\mathbf{u}_{\alpha^{AB}} \mathbf{u}_{\alpha^{AB}} - \frac{\mathbf{I}}{3} ds]\right),
\end{aligned}$$

$$\begin{aligned}
Q_C(\omega_C, \mathbf{M}) &= \frac{1}{V} \int D\mathbf{R}^C D\mathbf{u}_{\alpha^C} \delta[\mathbf{u}_{\alpha^C} - \frac{1}{c} \frac{d\mathbf{R}_{\alpha^C}}{ds}] \delta[|\mathbf{u}_{\alpha^C}| - 1] \\
&\times \exp\left(-\frac{\lambda_C}{2c} \int_0^{N_C} \left|\frac{d\mathbf{u}_{\alpha^C}}{ds}\right|^2 ds\right) \\
&\times \exp\left(-\int_0^{N_C} \{\omega_C - \mathbf{M} : [\mathbf{u}_{\alpha^C} \mathbf{u}_{\alpha^C} - \frac{\mathbf{I}}{3}]\} ds\right),
\end{aligned}$$

and

$$Q_{hA}(\omega_A) = \frac{1}{V} \int_V D\mathbf{R}^{hA} \exp\left(-\frac{3}{2a^2} \int_0^{N_{hA}} \left|\frac{d\mathbf{R}_{\alpha^{hA}}}{ds}\right|^2 ds - \int_0^{N_{hA}} \omega_A ds\right).$$

To calculate the single-chain partition functions, the positive propagator and inverse propagator of the polymer chain need to be defined. For instance,

$$\begin{aligned}
q_A^+(\mathbf{r}, s) &= \int_V D\mathbf{R}^B D\mathbf{u}_{\alpha^{AB}} D\mathbf{R}^A P^{AB}(\mathbf{R}^B, \mathbf{u}_{\alpha^{AB}}; [0, N_B]); \mathbf{R}^A; [0, s] \\
&\times \exp\left(-\int_0^{N_B} \{\omega_B - \mathbf{M} : [\mathbf{u}_{\alpha^{AB}} \mathbf{u}_{\alpha^{AB}} - \frac{\mathbf{I}}{3}]\} ds\right), \\
&\times \exp\left(-\int_0^s \omega_A ds'\right) \cdot \delta(\mathbf{r} - \mathbf{R}^A), \forall s \in [0, N_A],
\end{aligned} \tag{19}$$

$$\begin{aligned}
q_C^-(\mathbf{r}, \mathbf{u}, s) &= \int_V D\mathbf{R}^C D\mathbf{u}_{\alpha^C} P^C(\mathbf{R}^C, \mathbf{u}_{\alpha^C}; [0, N_C]) \\
&\times \exp\left(-\int_0^{N_C} \{-\mathbf{M} : [\mathbf{u}_{\alpha^C} \mathbf{u}_{\alpha^C} - \frac{\mathbf{I}}{3}]\} ds\right) \\
&\times \exp\left(-\int_0^s \omega_C ds'\right) \cdot \delta(\mathbf{r} - \mathbf{R}^C), \forall s \in [0, N_C].
\end{aligned} \tag{20}$$

The single-chain partition functions are obtained from the Feynman-Kac formulas (Freed (1972); Doi *et al.* (1988)). This yields the single-chain partition function

$$Q_{AB}(\omega_A, \omega_B, \mathbf{M}) = \frac{1}{V} \int_V d\mathbf{r} q_A^+(\mathbf{r}, N_A) \quad s \in [0, N_A]. \tag{21}$$

$$Q_{hA}(\omega_A) = \frac{1}{V} \int_V d\mathbf{r} q_A^h(\mathbf{r}, N_{hA}), \quad s \in [0, N_{hA}], \tag{22}$$

$$Q_C(\omega_C, \mathbf{M}) = \frac{1}{V} \int_V d\mathbf{r} \int_{S^2} d\mathbf{u} q_C(\mathbf{r}, \mathbf{u}, N_C), \quad \forall s \in [0, N_C]. \tag{23}$$

where the integrand function $q(\mathbf{r}, s)$ and $q(\mathbf{r}, \mathbf{u}, s)$ are the propagator. Physically $q(\mathbf{r}, s)$ is the probability that the s particle in the polymer chain at the \mathbf{r} position; $q(\mathbf{r}, \mathbf{u}, s)$ represents the probability that the s particle is at the \mathbf{r} position in the \mathbf{u} direction. These equations are solved by the modified diffusion equations (MDE) (Helfand (1975); Fredrickson *et al.* (2006)) in the presence of the mean fields

$$\frac{\partial}{\partial s} q_A^h(\mathbf{r}, s) = \left(\frac{a^2}{6} \nabla_{\mathbf{r}}^2 - \omega_A(\mathbf{r})\right) q_A^h(\mathbf{r}, s), \quad s \in (0, N_{hA}), \tag{24}$$

$$\frac{\partial}{\partial s} q_A^\pm(\mathbf{r}, s) = \left(\frac{a^2}{6} \nabla_{\mathbf{r}}^2 - \omega_A(\mathbf{r})\right) q_A^\pm(\mathbf{r}, s), \quad s \in (0, N_A), \tag{25}$$

$$\frac{\partial}{\partial s} q_B^\pm(\mathbf{r}, \mathbf{u}, s) = (\pm b \mathbf{u} \cdot \nabla_{\mathbf{r}} |_{\mathbf{u}} - \Gamma_B(\mathbf{r}, \mathbf{u})) q_B^\pm(\mathbf{r}, \mathbf{u}, s), \quad s \in (0, N_B), \tag{26}$$

$$\frac{\partial}{\partial s} q_C^\pm(\mathbf{r}, \mathbf{u}, s) = (\pm c \mathbf{u} \cdot \nabla_{\mathbf{r}} |_{\mathbf{u}} - \Gamma_C(\mathbf{r}, \mathbf{u})) q_C^\pm(\mathbf{r}, \mathbf{u}, s), \quad s \in (0, N_C). \tag{27}$$

with the initial conditions,

$$\begin{aligned}
q_A^-(\mathbf{r}, 0) &= q_A^h(\mathbf{r}, 0) = 1, \quad q_A^+(\mathbf{r}, 0) = \int_{S^2} d\mathbf{u} q_B^-(\mathbf{r}, \mathbf{u}, N_B), \\
q_B^+(\mathbf{r}, \mathbf{u}, 0) &= \frac{1}{4\pi} q_A^-(\mathbf{r}, N_A), \quad q_B^-(\mathbf{r}, \mathbf{u}, 0) = q_C^\pm(\mathbf{r}, \mathbf{u}, 0) = \frac{1}{4\pi}.
\end{aligned}$$

where $\Gamma_k(\mathbf{r}, \mathbf{u}) = \omega_k(\mathbf{r}) - \mathbf{M}(\mathbf{r}) : (\mathbf{u}\mathbf{u} - \frac{1}{3}\mathbf{I})$.

2 Self-consistent field equation

The Hamiltonian $H(\omega_i, \phi_i, \xi, \mathbf{M})$ of the system in equilibrium is needed in order to compute the partition function Z . We consider the variation of a first order of Hamiltonian with respect to the fields and density distributions. This yields

$$\frac{\delta H}{\delta \omega_A} = \rho_0 \left(-\phi_A - \frac{n_{AB}}{\rho_0 Q_{AB}} \frac{\delta Q_{AB}}{\delta \omega_A} - \frac{n_{hA}}{\rho_0 Q_{hA}} \frac{\delta Q_{hA}}{\delta \omega_A} \right) = 0, \quad (28)$$

$$\frac{\delta H}{\delta \omega_B} = \rho_0 \left(-\phi_B - \frac{n_{AB}}{\rho_0 Q_{AB}} \frac{\delta Q_{AB}}{\delta \omega_B} \right) = 0, \quad (29)$$

$$\frac{\delta H}{\delta \omega_C} = \rho_0 \left(-\phi_C - \frac{n_C}{\rho_0 Q_C} \frac{\delta Q_C}{\delta \omega_C} \right) = 0, \quad (30)$$

$$\frac{\delta H}{\delta \xi} = \rho_0 (\phi_A + \phi_B + \phi_C - 1) = 0, \quad (31)$$

$$\frac{\delta H}{\delta \phi_A} = \rho_0 (\chi_{AB} \phi_B + \chi_{AC} \phi_C - w_A - \xi) = 0, \quad (32)$$

$$\frac{\delta H}{\delta \phi_B} = \rho_0 (\chi_{AB} \phi_A + \chi_{BC} \phi_C - w_B - \xi) = 0, \quad (33)$$

$$\frac{\delta H}{\delta \phi_C} = \rho_0 (\chi_{AC} \phi_A + \chi_{BC} \phi_B - w_C - \xi) = 0, \quad (34)$$

$$\frac{\delta H}{\delta \mathbf{M}} = \rho_0 \left(\frac{1}{\eta} \mathbf{M} - \frac{n_{AB}}{\rho_0 Q_{AB}} \frac{\delta Q_{AB}}{\delta \mathbf{M}} - \frac{n_C}{\rho_0 Q_C} \frac{\delta Q_C}{\delta \mathbf{M}} \right) = 0. \quad (35)$$

The density distributions and orientational order parameter is defined as

$$\begin{aligned} \phi_A(\mathbf{r}) &= \phi_A^h(\mathbf{r}) + \phi_A^c(\mathbf{r}) := -\frac{n_{hA}}{\rho_0 Q_{hA}} \frac{\delta Q_{hA}}{\delta \omega_A} - \frac{n_{AB}}{\rho_0 Q_{AB}} \frac{\delta Q_{AB}}{\delta \omega_A} \\ &= \frac{n_{hA}}{\rho_0 V Q_{hA}} \int_0^{N_{hA}} ds q_A^h(\mathbf{r}, s) q_A^h(\mathbf{r}, N_{hA} - s) + \frac{n_{AB}}{\rho_0 V Q_{AB}} \int_0^{N_A} ds q_A^-(\mathbf{r}, s) q_A^+(\mathbf{r}, N_A - s), \end{aligned} \quad (36)$$

$$\phi_B(\mathbf{r}) := -\frac{4\pi n_{AB}}{\rho_0 Q_{AB}} \frac{\delta Q_{AB}}{\delta \omega_B} = \frac{4\pi n_{AB}}{\rho_0 V Q_{AB}} \int_0^{N_B} ds \int d\mathbf{u} q_B^-(\mathbf{r}, \mathbf{u}, s) q_B^+(\mathbf{r}, \mathbf{u}, N_B - s), \quad (37)$$

$$\phi_C(\mathbf{r}) := -\frac{4\pi n_C}{\rho_0 Q_C} \frac{\delta Q_C}{\delta \omega_C} = \frac{4\pi n_C}{\rho_0 V Q_C} \int_0^{N_C} ds \int d\mathbf{u} q_C^-(\mathbf{r}, \mathbf{u}, s) q_C^+(\mathbf{r}, \mathbf{u}, N_C - s), \quad (38)$$

$$\mathbf{S}_B(\mathbf{r}) := \frac{4\pi n_{AB}}{\rho_0 Q_{AB}} \frac{\delta Q_{AB}}{\delta \mathbf{M}} = \frac{4\pi n_{AB}}{\rho_0 V Q_{AB}} \int_0^{N_B} ds \int d\mathbf{u} q_B^-(\mathbf{r}, \mathbf{u}, s) \left(\mathbf{u} \mathbf{u} - \frac{\mathbf{I}}{3} \right) q_B^+(\mathbf{r}, \mathbf{u}, N_B - s), \quad (39)$$

$$\mathbf{S}_C(\mathbf{r}) := \frac{4\pi n_C}{\rho_0 Q_C} \frac{\delta Q_C}{\delta \mathbf{M}} = \frac{4\pi n_C}{\rho_0 V Q_C} \int_0^{N_C} ds \int d\mathbf{u} q_C^-(\mathbf{r}, \mathbf{u}, s) \left(\mathbf{u} \mathbf{u} - \frac{\mathbf{I}}{3} \right) q_C^+(\mathbf{r}, \mathbf{u}, N_C - s). \quad (40)$$

The self-consistent field equations are

$$\begin{cases} w_A(\mathbf{r}) = \chi_{AB} \phi_B(\mathbf{r}) + \chi_{AC} \phi_C(\mathbf{r}) - \xi(\mathbf{r}), \\ w_B(\mathbf{r}) = \chi_{AB} \phi_A(\mathbf{r}) + \chi_{BC} \phi_C(\mathbf{r}) - \xi(\mathbf{r}), \\ w_C(\mathbf{r}) = \chi_{AC} \phi_A(\mathbf{r}) + \chi_{BC} \phi_B(\mathbf{r}) - \xi(\mathbf{r}), \\ 1 = \phi_A(\mathbf{r}) + \phi_B(\mathbf{r}) + \phi_C(\mathbf{r}), \\ \mathbf{M}(\mathbf{r}) = \eta (\mathbf{S}_B(\mathbf{r}) + \mathbf{S}_C(\mathbf{r})). \end{cases} \quad (41)$$

When Eq.(36)-(40) satisfy Eq.(41), the effective Hamiltonian $H(\omega_i^*, \phi_i^*, \xi^*, \mathbf{M}^*)$ is obtained at the saddle point $(\omega_i^*, \phi_i^*, \xi^*, \mathbf{M}^*)$.

The partition function Z given by Eq.(17), can be approximated (Matsen (1995); Hong and Noolandi (1981)) $\exp(-H(\omega_i^*, \phi_i^*, \xi^*, \mathbf{M}^*))$.

This yields

$$\mathcal{F} = -\kappa_B T \ln Z \approx \kappa_B T H(\omega_i^*, \phi_i^*, \xi^*, \mathbf{M}^*). \quad (42)$$

3 Dimensionless model

We use a dimensionless model in order to effectively simulate the bilayer membrane and obtain the key parameters. We denote the number of molecules as $n_{AB} = \tilde{n}_{AB}n = 1 \cdot n$, $\tilde{n}_{hA} = \tilde{n}_{hA}/n$, $\tilde{n}_C = \tilde{n}_C/n$, and redefine $\omega'_A = \omega_A N$, $\omega'_B = \omega_B N$, and $\omega'_C = \omega_C N$ as ω_A , ω_B and ω_C . We write $\frac{\lambda_B}{bN}$, and $\frac{\lambda_C}{cN}$ as λ_B and λ_C respectively. $\beta_B = L_B/R_g$ and $\beta_C = L_C/R_g$ are the geometrical asymmetry parameters (Tao *et al.* (2007); Li *et al.* (2013)) for the rod length $L_B = bN$ and $L_C = cN$. We write $f_A = N_A/N$, $f_B = 1 - f_A$, $f_C = N_C/N$, and $f_h = N_h/N$; these volume fractions provide normalized volumes of the polymer. The Flory-Huggins interaction parameters $\chi_{AB}N$, $\chi_{AC}N$ and $\chi_{BC}N$ describe the molecular chain interactions, and the Maier-Suape interaction parameter ηN controls the orientational strength of rigid molecules. The free energy per chain of the normalized mixed system of AB diblock copolymers and blend homopolymers with volume V in the canonical ensemble is

$$\begin{aligned} \frac{N\mathcal{F}}{\rho_0 V} = & \frac{1}{V} \int_V d\mathbf{r} \left\{ \sum_{i \neq j} \chi_{ij} N \phi_i \phi_j - \sum_i \omega_i \phi_i + \frac{1}{2\eta N} \mathbf{M} : \mathbf{M} - \zeta \left(\sum_i \phi_i - 1 \right) \right\} - \frac{1}{\tilde{\rho}} \log(Q_{AB}^{n_{AB}} Q_{hA}^{n_{hA}} Q_C^{n_C}) \\ & - \frac{1}{\tilde{\rho}} \log\left(\left(\frac{Ne}{\rho_0}\right)^{\tilde{n}_{AB} + \tilde{n}_{hA} + \tilde{n}_C}\right) + \frac{N}{2\tilde{\rho}} \mathbf{E} \mathbf{f} \mathbf{n}^T. \end{aligned} \quad (43)$$

where $\tilde{\rho} = 1 + \tilde{n}_h f_h + \tilde{n}_C f_C$, $\mathbf{E} = (e_{AA}, e_{AA}, e_{BB}, e_{CC})$, $\mathbf{f} = \text{diag}(f_{hA}, f_A, f_B, f_C)$, and $\mathbf{n} = (\tilde{n}_{hA}, \tilde{n}_{AB}, \tilde{n}_{AB}, \tilde{n}_C)$. The single-chain partition can now be written as as

$$Q_{AB}[\omega_A(\mathbf{r}), \omega_B(\mathbf{r}), \mathbf{M}(\mathbf{r})] = \frac{\tilde{\rho}}{V} \int_V d\mathbf{r} q_A^+(\mathbf{r}, f_A), \quad (44)$$

$$Q_{hA}[\omega_A(\mathbf{r})] = \frac{\tilde{\rho}}{\tilde{n}_{hA} V} \int_V d\mathbf{r} q_A^h(\mathbf{r}, f_{hA}), \quad (45)$$

$$Q_C[\omega_C(\mathbf{r}), \mathbf{M}(\mathbf{r})] = \frac{\tilde{\rho}}{\tilde{n}_C V} \int_V d\mathbf{r} q_C^+(\mathbf{r}, \mathbf{u}, f_C), \quad (46)$$

the MEDs become

$$\frac{\partial}{\partial s} q_A(\mathbf{r}, s) = (R_g^2 \nabla_{\mathbf{r}}^2 - \omega_A(\mathbf{r})) q_A(\mathbf{r}, s), \quad q_A \in \{q_A^h, q_A^\pm\}, \quad (47)$$

$$\frac{\partial}{\partial s} q_k^\pm(\mathbf{r}, \mathbf{u}, s) = (\pm \beta_k \mathbf{u} \cdot \nabla_{\mathbf{r}} - \Gamma_k(\mathbf{r}, \mathbf{u})) q_k^\pm(\mathbf{r}, \mathbf{u}, s), \quad k \in \{B, C\}, \quad (48)$$

with initial conditions,

$$\begin{aligned} q_A^-(\mathbf{r}, 0) = q_A^h(\mathbf{r}, 0) = 1, \quad q_A^+(\mathbf{r}, 0) = \int_{S^2} d\mathbf{u} q_B^-(\mathbf{r}, \mathbf{u}, f_B), \\ q_B^+(\mathbf{r}, \mathbf{u}, 0) = \frac{1}{4\pi} q_A^-(\mathbf{r}, f_A), \quad q_B^-(\mathbf{r}, \mathbf{u}, 0) = q_C^\pm(\mathbf{r}, \mathbf{u}, 0) = \frac{1}{4\pi}. \end{aligned}$$

The density distributions and orientational order parameters are

$$\phi_A(\mathbf{r}) = \phi_A^h(\mathbf{r}) + \phi_A^c(\mathbf{r}) = \frac{1}{Q_{hA}} \int_0^{f_{hA}} ds q_A^h(\mathbf{r}, s) q_A^h(\mathbf{r}, f_{hA} - s) + \frac{1}{Q_{AB}} \int_0^{f_A} ds q_A^-(\mathbf{r}, s) q_A^+(\mathbf{r}, f_A - s), \quad (49)$$

$$\phi_k(\mathbf{r}) = \frac{4\pi}{Q_k} \int_0^{f_k} ds \int d\mathbf{u} q_k^-(\mathbf{r}, \mathbf{u}, s) q_k^+(\mathbf{r}, \mathbf{u}, f_k - s), \quad k \in \{B, C\}, \quad (50)$$

$$\mathbf{S}(\mathbf{r}) = \mathbf{S}_B(\mathbf{r}) + \mathbf{S}_C(\mathbf{r}) = \sum_{k \in \{B, C\}} \frac{4\pi}{Q_k} \int d\mathbf{u} \int_0^{f_k} ds (\mathbf{u} \mathbf{u} - \frac{\mathbf{I}}{3}) q_k^-(\mathbf{r}, \mathbf{u}, s) q_k^+(\mathbf{r}, \mathbf{u}, f_k - s). \quad (51)$$

The self-consistent field equations are

$$\begin{cases} \omega_A(\mathbf{r}) = \chi_{AB} N \phi_B(\mathbf{r}) + \chi_{AC} N \phi_C(\mathbf{r}) - \zeta(\mathbf{r}), \\ \omega_B(\mathbf{r}) = \chi_{AB} N \phi_A(\mathbf{r}) + \chi_{BC} N \phi_C(\mathbf{r}) - \zeta(\mathbf{r}), \\ \omega_C(\mathbf{r}) = \chi_{AC} N \phi_A(\mathbf{r}) + \chi_{BC} N \phi_B(\mathbf{r}) - \zeta(\mathbf{r}), \\ 1 = \phi_A(\mathbf{r}) + \phi_B(\mathbf{r}) + \phi_C(\mathbf{r}), \\ \mathbf{M}(\mathbf{r}) = \eta \mathbf{N} \mathbf{S}(\mathbf{r}). \end{cases} \quad (52)$$

4 Numerical method

We now introduce the numerical method to solve the SCF equations. Generally, SCFT models can be solved by iteration. In solving SCF equations, we first need to solve the MDEs. This is the most time-consuming step. After solving the SCF equations, the free energy and structure of the bilayers are obtained, allowing the chemical potential of C polymers to be calculated. A flowchart of the procedure is shown in Figure 2.

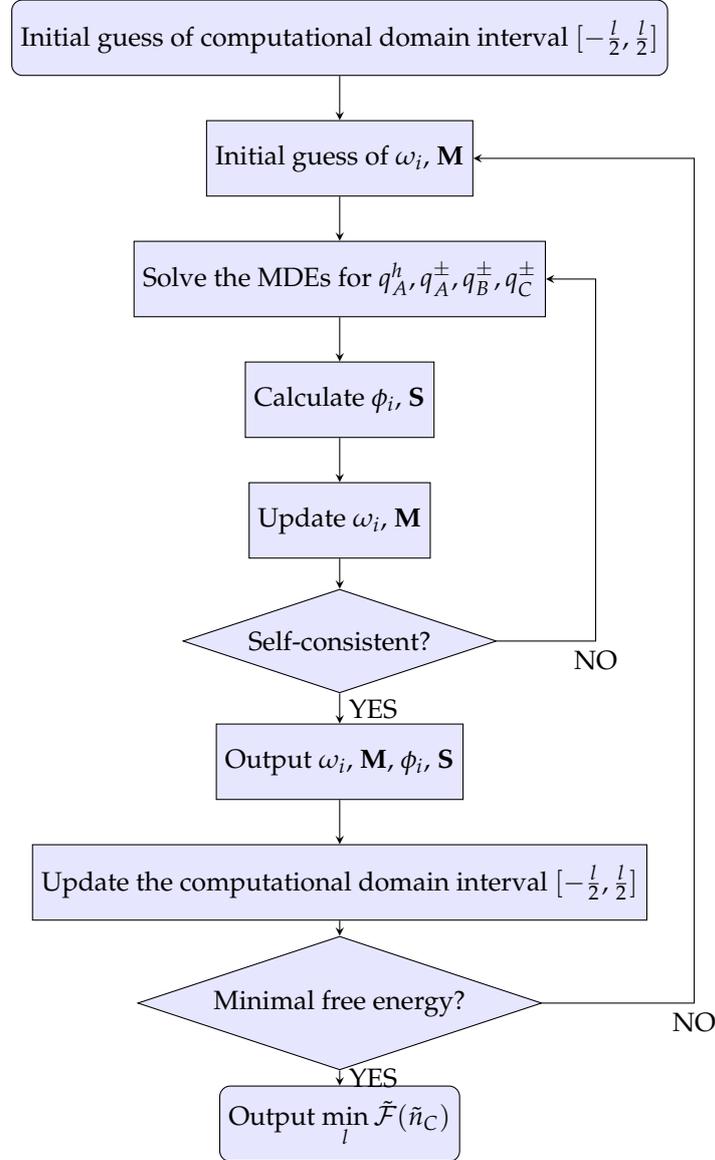


Figure 2: Flowchart for solving the normalized one-dimensional SCF in the canonical ensemble.

4.1 Solving the MDEs

We illustrate our procedure for the more complicated Eq. (48); the simpler Eq. (47) is also solved by this approach. We reformulate Eq. (48) with an initial value q^0 and a reflection boundary condition:

$$\frac{\partial}{\partial s} q(x, \mathbf{u}, s) = a \frac{\partial}{\partial x} q(x, \mathbf{u}, s) - \Gamma(x, \mathbf{u}) q(x, \mathbf{u}, s), \quad (53)$$

$$q(x, \mathbf{u}, 0) = q^0(x, \mathbf{u}), \quad \frac{\partial}{\partial x} q(x_{in}, \mathbf{u}, s) = 0, \quad (54)$$

The computational domain is truncated to the optimal interval $[-l^*/2, l^*/2]$ with length l^* . This minimizes the free energy and places the boundary condition at $x_{in} = \pm l^*$. (The sign + or - is determined by the sign of the coefficient a). We employ, the well-known Strang splitting method (Strang (1968)) and obtain the following second-order semi-discrete scheme for (53) with step size Δs ,

$$\begin{aligned} u^{k+\frac{1}{2}}(x, \mathbf{u}) &= \exp(-\tilde{\Gamma}(x, \mathbf{u})\frac{\Delta s}{2})u^k(x, \mathbf{u}), \\ u^*(x, \mathbf{u}) &= \exp(\tilde{a}(x)\Delta s\partial_x)u^{k+\frac{1}{2}}(x, \mathbf{u}), \\ u^{k+1}(x, \mathbf{u}) &= \exp(-\tilde{\Gamma}_0(x, \mathbf{u})\frac{\Delta s}{2})u^*(x, \mathbf{u}), \end{aligned}$$

where $u^k(x, \mathbf{u})$ is the numerical approximation of $u(x, \mathbf{u}, s_k)$, and $s_k = k\Delta s$. The solution for $u^*(x, \mathbf{u})$ is obtained by the well-known Crank-Nicolson scheme (Morton and Mayers (2005)).

4.2 Solve the SCF equations

The SCF equations (52) are solved by Picard iteration, where the fields $\omega_i^{new}(x)$, $\mathbf{M}^{new}(x)$ are updated from an old set of fields, $\omega_i^{old}(x)$, $\mathbf{M}^{old}(x)$. In a bit more detail, we first solve the MDEs with these old mean fields to obtain propagators, $q_A^h, q_A^\pm, q_B^\pm, q_C^\pm$, which are directly used to calculate $\phi_i(x)$, and $\mathbf{S}(x)$. Next, we assign Lagrange multiplier $\zeta(x)$ according to Eq. (41). These fields ω_i , \mathbf{M} are updated according to Eq. (55)-(58). These steps are repeated until the errors of SCF equations are sufficiently small. The free energy is then obtained by Eq. (43).

The old fields $\omega_i^{old}(x)$, $\mathbf{M}^{old}(x)$ (and its corresponding concentrations $\phi_i^{old}(x)$, $\mathbf{S}^{old}(x)$) are updated by using the iteration,

$$\omega_A^{new}(x) = (1 - \alpha_1)\omega_A^{old}(x) + \alpha_1 \left[\chi_{AB}N\phi_B^{old}(x) + \chi_{AC}N\phi_C^{old}(x) - \zeta^{old}(x) \right], \quad (55)$$

$$\omega_B^{new}(x) = (1 - \alpha_1)\omega_B^{old}(x) + \alpha_1 \left[\chi_{AB}N\phi_A^{old}(x) + \chi_{BC}N\phi_C^{old}(x) - \zeta^{old}(x) \right], \quad (56)$$

$$\omega_C^{new}(x) = (1 - \alpha_1)\omega_C^{old}(x) + \alpha_1 \left[\chi_{AC}N\phi_A^{old}(x) + \chi_{BC}N\phi_B^{old}(x) - \zeta^{old}(x) \right], \quad (57)$$

$$\mathbf{M}^{new}(\mathbf{r}) = (1 - \alpha_2)\mathbf{M}^{old}(\mathbf{r}) + \alpha_2\eta\mathbf{N}\mathbf{S}^{old}(\mathbf{r}). \quad (58)$$

where α_i , $i = 1, 2$, are update ratios which, in our calculations, are chosen as $\alpha_1 = 0.03$ and $\alpha_2 = 0.01$. This Picard-type iteration is robust, but converges slowly. Therefore when the fields require an inordinately large number of Picard iterations in order to reduce the errors of updated fields to less than 0.1, we speed up convergence by applying Anderson acceleration (Thompson *et al.* (2004)) to the corresponding fixed point problem given by Eq. (41). In both the Picard and Anderson iterations, the Lagrange multiplier $\zeta^{new}(x)$ is

$$\zeta^{new}(x) = \frac{1}{3} \left(\chi_{AB}N(1 - \phi_C^{old}(x)) + \chi_{AC}N(1 - \phi_B^{old}(x)) + \chi_{BC}N(1 - \phi_A^{old}(x)) - \omega_A^{old}(x) - \omega_B^{old}(x) - \omega_C^{old}(x) \right).$$

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