Supplementary materials

Coarse-grained (CG) DOPE lipid

The CG DOPE lipid [1,2] is represented by 14 beads: two for headgroups, two for glycerol groups and five for each fatty acid tail, as shown in Fig. A1.

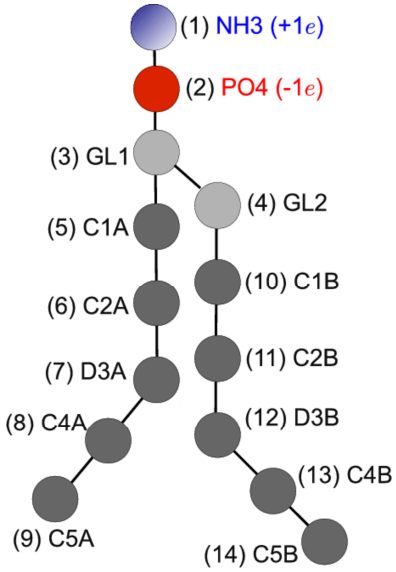

Fig. A1 Schematic show of the CG DOPE lipid, where all CG atoms except for NH3 and PO4 are free of charge.

The length-dependent bond energy is calculated by $U_{bond} = \frac{1}{2} K_{bond} (R - R_0)^2$. K_{bond} = 1250 kJ/mol for all bonds and R_0 = 0.47 nm for nearly all bonds except for the bond of GL1-GL2, which is 0.37 nm. The angle-dependent bond energy is calculated by $U_{angle}(\theta) = \frac{1}{2} K_{angle} \{\cos(\theta) - \cos(\theta_0)\}^2$. K_{angle} = 25 kJ/mol for nearly all angles except for C2A-D3A-C4A and C2B-D3B-C4B, which are 45 kJ/mol. θ0 = 180 degrees for nearly all angles except for PO4-GL1-GL2, C2A-D3A-C4A and C2B-D3B-C4B, θ_0= 120 degrees. The used Lennard-Jones (LJ) potential is given by

$U_{LJ}(r_{ij}) = 4\varepsilon\left[\left(\frac{\sigma}{r_{ij}}\right)^{12} - \left(\frac{\sigma}{r_{ij}}\right)^{6}\right]$, where σ=0.47 nm for all particles in the present simulations. According to MARTINI force field [1, 2], particles in the present simulations can be classified into six groups (Fig. A1): Qd (NH3); Qa (PO4); Na (GL1 and GL2), C1 (C1A, C2A, C4A, C5A, C1B, C2B, C4B and C5B), C_3 (D3A and D3B) and POL (W in water in Fig. A2). Accordingly, there are eight levels of ε in the present simulations: O (Q_d-N_a), ε = 5.6 kJ/mol; I (POL-Qd, POL-Qa), ε = 5.0 kJ/mol; II (Na-Qa), ε = 4.5 kJ/mol; III (Qa-Qd, Na-N_a, POL-N_a), ε = 4.0 kJ/mol; IV (Q_d-Q_d, Q_a-Q_a, C1-C_1, C1-C_3, C_3-C_3), ε = 3.5 kJ/mol; VI (Qd-C_3, Q_a-C_3, Na-C_1, Na-C_3, POL-C3), ε = 2.7 kJ/mol; VII (Qd-C_1, Q_a-C_1), ε = 2.3 kJ/mol and VIII (POL-C1), ε = 2.0 kJ/mol. The electrostatic potential energy is calculated with $U_{el}(r_{ij}) = \frac{q_i q_j}{4\pi\varepsilon_0\varepsilon_r r_{ij}}$, where $\varepsilon_0 = 8.85\times10^{-12}$ F/m is the vacuum permittivity and the relative permittivity ε_r = 2.5 is used in the present simulations.

CG Polarizable CG water molecule

Figure A2 shows the used CG polarizable water molecule [1], which is represented by W, WP and WM beads. W is charge-free and interacts with other CG water molecules and lipids via the LJ potential with ε = 4.0 kJ/mol (Level III) and σ = 0.47 nm. WP and WM are bonded to W with a fixed bond length of 0.14 nm and have charge +0.46e and -0.46e respectively. They interact with other particles through electrostatic interactions. The bond angle of WP-W-WM is flexible and constrained by a harmonic potential of $U_{angle}(\theta) = \frac{1}{2}K_{angle}\{\cos(\theta) - \cos(\theta_0)\}^2$, here $\theta_0 = 0$ and k_{angle} = 4.2 kJ/(mol·rad^2), indicating that the dipole of water is 0 C·m when there is no external electric field and can be polarized by external electric fields due to the angle change. One CG water molecule represents a cluster of four real water molecules.

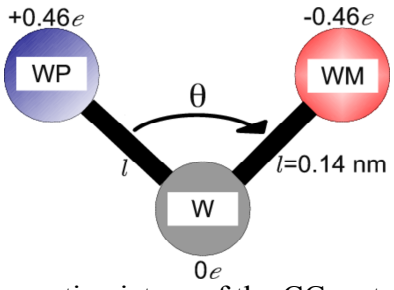

Fig.A2: Schematic picture of the CG water molecule.

Electronic Supplementary Material (ESI) for Soft Matter

The larger system with 3200 DOPE lipids and 28800 water molecules

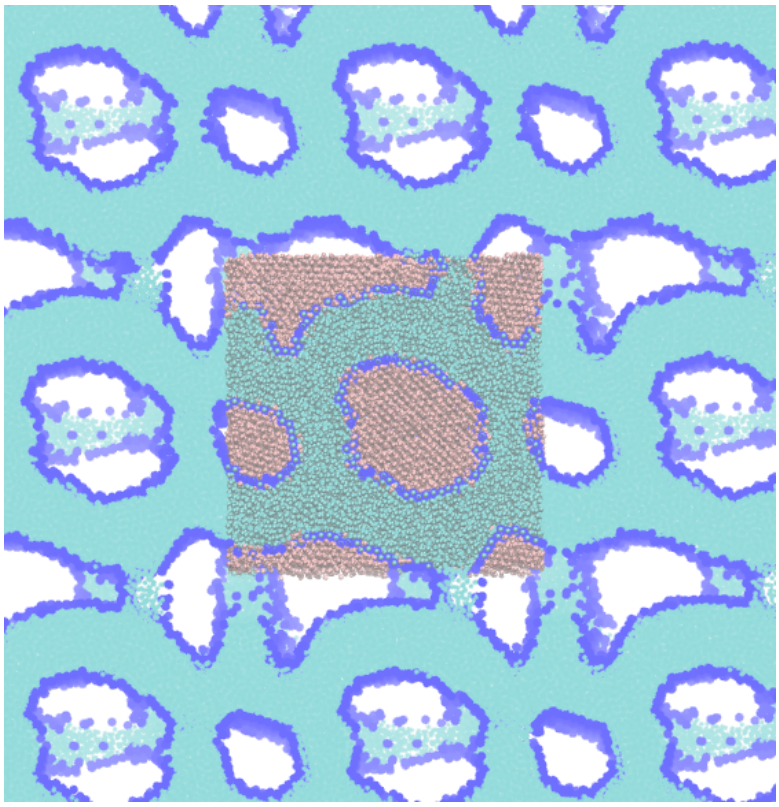

Fig.A3:Snapshots of the larger system with 3200 DOPE lipids and 28800 water molecules (36 w/l) in external electric field of 0.5 V/nm at 50 ns. The direction of the external electric field is perpendicular to the paper and points outside. Water is removed for clarity in the images of the primary simulation cell. PO4 and NH3 of DOPE lipids are shown in blue to indicate the interfaces of water/lipids and other particles of lipids are shown in cyan. Water molecules in the primary simulation cell are shown in red and the solid balls indicates the primary simulation cell.

Movies

Movie1: Self-assembly in the 24 w/l system without external electric field. CG water molecules are shown in red balls and lipid headgroups (NO3 and PO4 atoms in Fig. A1) in blue balls. Lipid tails are shown in transparent gray balls for clarity. For clarity, the system rotates around the y axis in the movie.

Movie2: Self-assembly in the 24 w/l system under the 0.5V/nm electric field. The direction of the external electric field is along the *z* direction. The representation and color schema are the same with that in Movie1.

Movie3: The external electric field was removed at the end of Movie2. The representation and color schema are the same with that in Movie1.

References

[1] Yesylevskyy S. O., Schäfer L. V., Sengupta D., Marrink S. J, *PLoS Comput. Biol.* **2010**, *6*, e1000810.

[2] Marrink S. J., Risselada H. J., Yefimov S.,. Tieleman D. P, de Vries A. H., *J. Phys. Chem. B* **2007**, *111*, 7812.