Supplementary information

Modeling the Hydrophobicity of Nanoparticles and their Interaction with Lipids and Proteins

Ali Ramazani[‡], Taraknath Mandal[‡], Ronald G Larson*
Department of Chemical Engineering, University of Michigan, 2300 Hayward St., Ann Arbor, USA

Table S1: LJ parameters for self-interaction and cross-interaction between new Martini bead type (C6) and conventional Martini bead types

Bead Type	C_6	C_{12}
C1 – C6	0.12935	0.001394325
C2 – C6	0.13366	0.0014408
C3 – C6	0.15091	0.0016267
C4 – C6	0.15091	0.0016267
C5 – C6	0.15091	0.0016267
C6 – C6	0.15091	0.0016267
N0 – C6	0.15091	0.0016267
Na – C6	0.15091	0.0016267
Nd – C6	0.15091	0.0016267
Nda – C6	0.15091	0.0016267
P1 – C6	0.1562975	0.001684775
P2 – C6	0.1562975	0.001684775
P3 – C6	0.15091	0.0016267
P4 – C6	0.1379725	0.001487275
P5 – C6	0.1379725	0.001487275
Q0 – C6	0.1379725	0.001487275
Qa – C6	0.1379725	0.001487275
Qd – C6	0.1379725	0.001487275
Gda – C6	0.1379725	0.001487275
SP4 – C6	0.1379725	0.001487275
SP1 – C6	0.1562975	0.001684775
SNd – C6	0.15091	0.0016267
SNa – C6	0.15091	0.0016267
SC5 – C6	0.15091	0.0016267
SC4 – C6	0.15091	0.0016267
SC3 – C6	0.15091	0.0016267
SC2 – C6	0.13366	0.0014408
SC1 – C6	0.12935	0.001394325
AC1 – C6	0.12935	0.001394325
AC2 – C6	0.13366	0.0014408
BP4 – C6	0.1379725	0.001487275

Table S2: LJ parameters for self-interaction and cross-interaction between new Martini bead type (C7) and conventional Martini bead types

Bead Type	C_6	C_{12}
C1 – C7	0.12504	0.00134785
C2 – C7	0.13366	0.0014408
C3 – C7	0.15091	0.0016267
C4 – C7	0.15091	0.0016267
C5 – C7	0.15091	0.0016267
C8 – C7	0.15091	0.0016267
N0 – C7	0.15091	0.0016267
Na – C7	0.15091	0.0016267
Nd – C7	0.15091	0.0016267
Nda – C7	0.15091	0.0016267
P1 – C7	0.161685	0.00174285
P2 – C7	0.161685	0.00174285
P3 – C7	0.15091	0.0016267
P4 – C7	0.142285	0.00153375
P5 – C7	0.142285	0.00153375
Q0 – C7	0.142285	0.00153375
Qa – C7	0.142285	0.00153375
Qd – C7	0.142285	0.00153375
Gda – C7	0.142285	0.00153375
SP4 – C7	0.142285	0.00153375
SP1 – C7	0.161685	0.00174285
SNd – C7	0.15091	0.0016267
SNa – C7	0.15091	0.0016267
SC5 – C7	0.15091	0.0016267
SC4 – C7	0.15091	0.0016267
SC3 – C7	0.15091	0.0016267
SC2 – C7	0.13366	0.0014408
SC1 – C7	0.12504	0.00134785
AC1 – C7	0.12504	0.00134785
AC2 – C7	0.13366	0.0014408
BP4 – C7	0.142285	0.00153375

Table S3: LJ parameters for self-interaction and cross-interaction between new Martini bead type (C8) and conventional Martini bead types

Bead Type	C_6	C_{12}
C1 – C8	0.12073	0.001301375
C2 – C8	0.13366	0.0014408
C3 – C8	0.15091	0.0016267
C4 – C8	0.15091	0.0016267
C5 – C8	0.15091	0.0016267
C8 – C8	0.15091	0.0016267
N0 – C8	0.15091	0.0016267
Na – C8	0.15091	0.0016267
Nd – C8	0.15091	0.0016267
Nda – C8	0.15091	0.0016267
P1 – C8	0.1670725	0.001800925
P2 – C8	0.1670725	0.001800925
P3 – C8	0.15091	0.0016267
P4 – C8	0.1465975	0.001580225
P5 – C8	0.1465975	0.001580225
Q0 – C8	0.1465975	0.001580225
Qa – C8	0.1465975	0.001580225
Qd – C8	0.1465975	0.001580225
Gda – C8	0.1465975	0.001580225
SP4 – C8	0.1465975	0.001580225
SP1 – C8	0.1670725	0.001800925
SNd – C8	0.15091	0.0016267
SNa – C8	0.15091	0.0016267
SC5 – C8	0.15091	0.0016267
SC4 – C8	0.15091	0.0016267
SC3 – C8	0.15091	0.0016267
SC2 – C8	0.13366	0.0014408
SC1 – C8	0.12073	0.001301375
AC1 – C8	0.12073	0.001301375
AC2 – C8	0.13366	0.0014408
BP4 – C8	0.1465975	0.001580225

Interfacial Surface tension calculation: The interfacial surface tension was computed using the following equation:

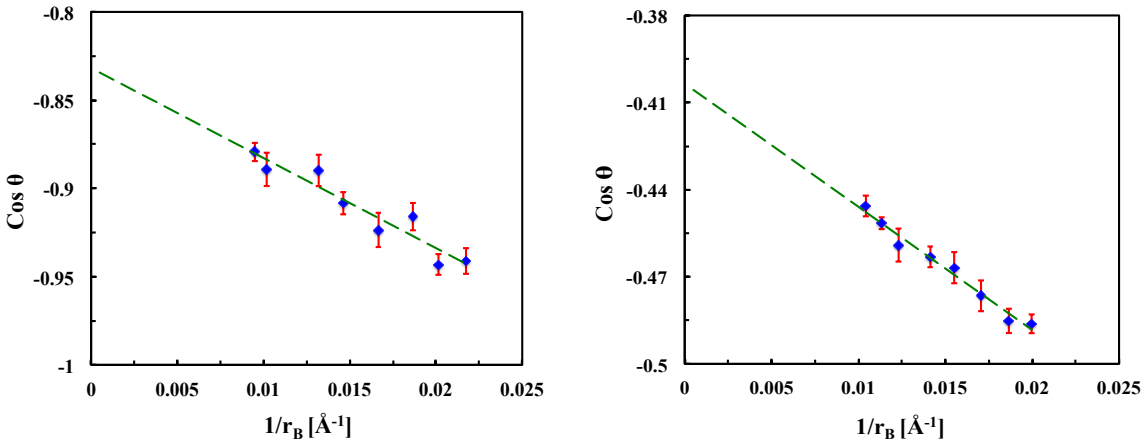
$$\gamma = \frac{1}{2} L_Z \left(< Pz - \frac{P_X + P_Y}{2} >\right)$$

Here P_Z is the average pressure along the perpendicular direction of the interface. P_X and P_Y are the average pressure along the two parallel directions of the interface. L_Z is the box length along the perpendicular direction of the interface. First, we computed the surface tension of the MARTINI water/air interface. The simulation box dimensions were taken to be 3.64 x 3.64 x 13.64 nm, which contained 400 water beads. The water/air surface tension was found to be 29.7 mN/M. To verify the dependency of the number of beads in the water slab on the interfacial surface tension, we also computed the surface tension with the slabs containing 800 (box 3.64 x 3.64 x 17 nm) and 1696 water beads (box 3.64 x 3.64 x 26 nm). The surface tension values were obtained as 29.4 and 29.5 mN/M for 800 beads slab and 1696 beads slab, respectively. Thus we obtained a constant water/air surface tension irrespective of the number of water beads in the water slab. Similarly, we computed the dodecane/air and dodecane/water surface tensions using MARTINI, polarizable MARTINI and BMW-MARTINI model. Each simulation ran for 1000 ns. The values are presented in **Table S4.**

Table S4: Surface tension values of different interfaces.

Interface/Model	MARTINI	Polarizable MARTINI	BMW-MARTINI	Experimental value
Water/air (mN/m)	29.7 (400 beads) 29.4 (800 beads) 29.5 (1696 beads)	30.9* 34.9# (1824 water molecules)	77.8 (488 water molecules)	73
Dodecane/air (mN/m)	22.8 (204 dodecane molecules)	22.8 (204 dodecane molecules)	22.8 (204 dodecane molecules)	24
Water/dodecane (mN/m)	40.6 (204 dodecane + 666 water beads) 40.8 (408 dodecane + 1639 water beads)	51.9 (204 dodecane + 812 water molecules)	93.6 (204 dodecane + 1490 water molecules) 93.1 (408 dodecane + 2720 water molecules)	52

*Shift and #PME method was used for electrostatic energy computation.



(a) (b)

Figure S1: Cosine of the contact angle of BMW-MARTINI water as a function of the droplet base curvature for (111) FCC surface composed of beads of type (a) C1 and (b) P1.

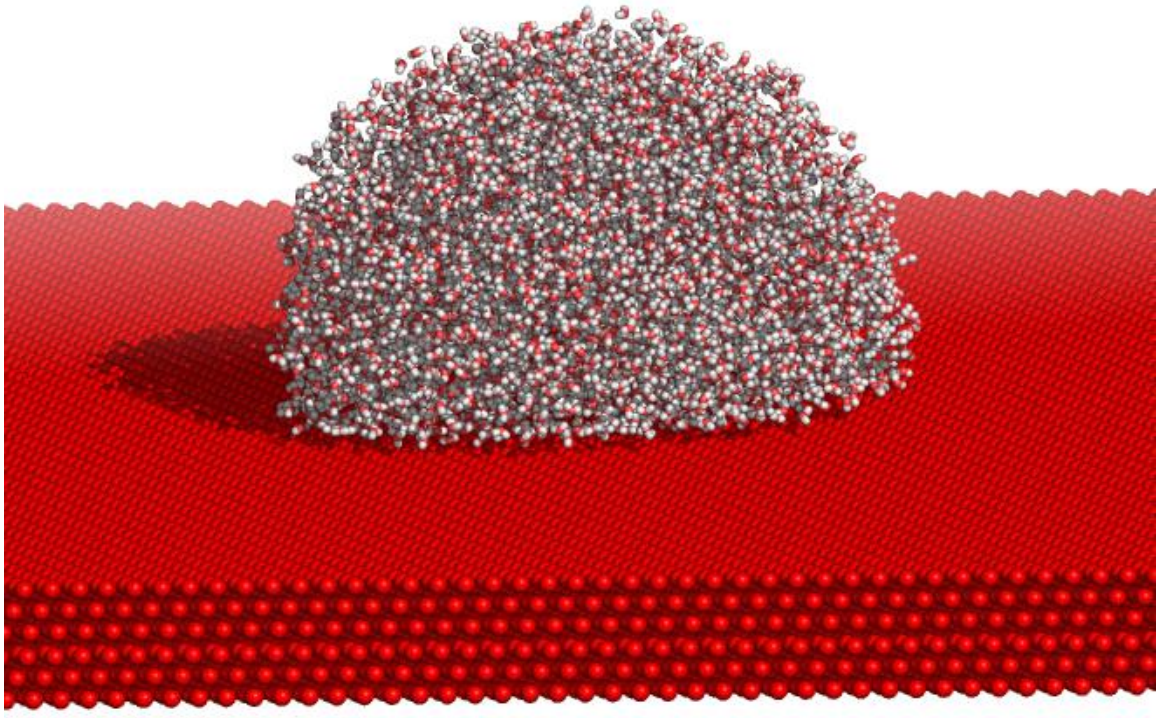


Figure S2: Snapshot of a BMW-MARTINI water droplet on top of a P4 FCC surface obtained after 140 ns-long MD simulation.

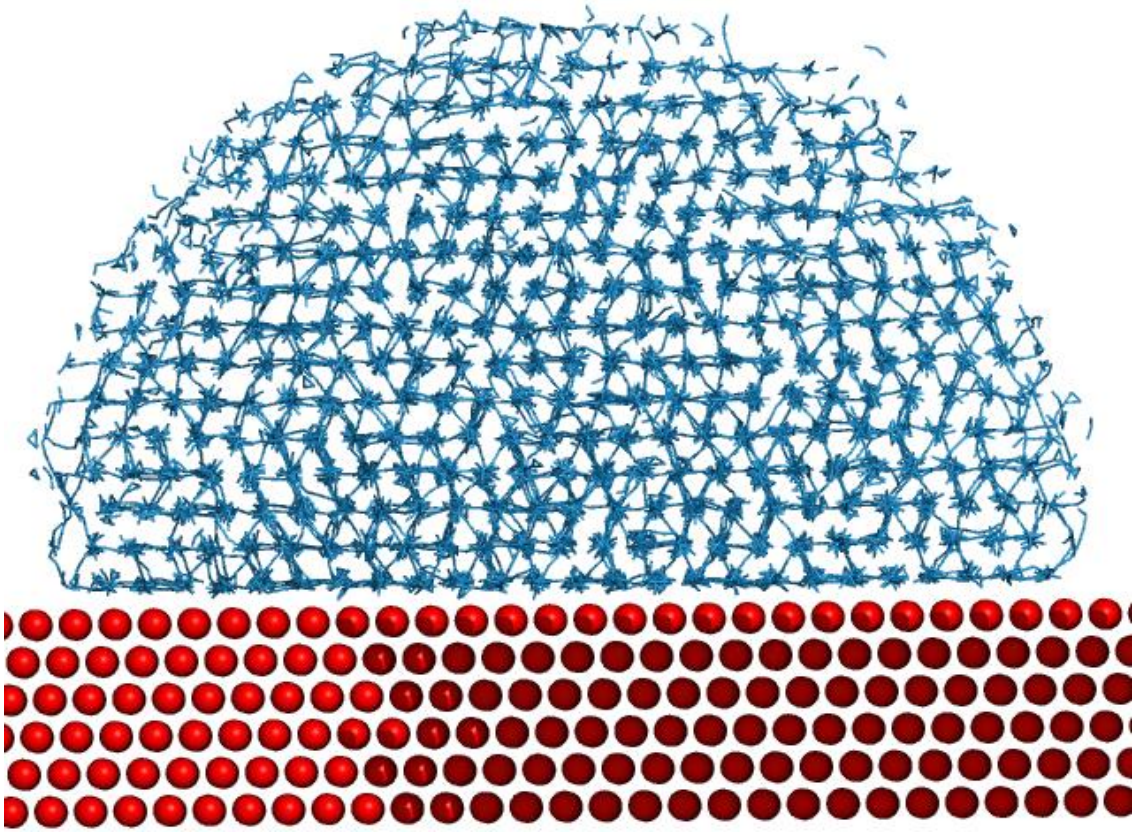


Figure S3: Snapshot of the crystalline polarizable MARTINI water droplet (blue) top of a FCC crystal surface (red). The crystal surface and the water molecules are shown with 'VDW sphere' and 'line' representation, respectively. We carried out the contact angle calculations with these kinds of frozen topologies. The polarizable MARTINI water contact angles are 121° and 30° for the C1 and N0 bead, respectively, which are close to the corresponding MARTINI values. However, the contact angles may not be reliable, as the droplets might not have adapted the correct topology before they were frozen.

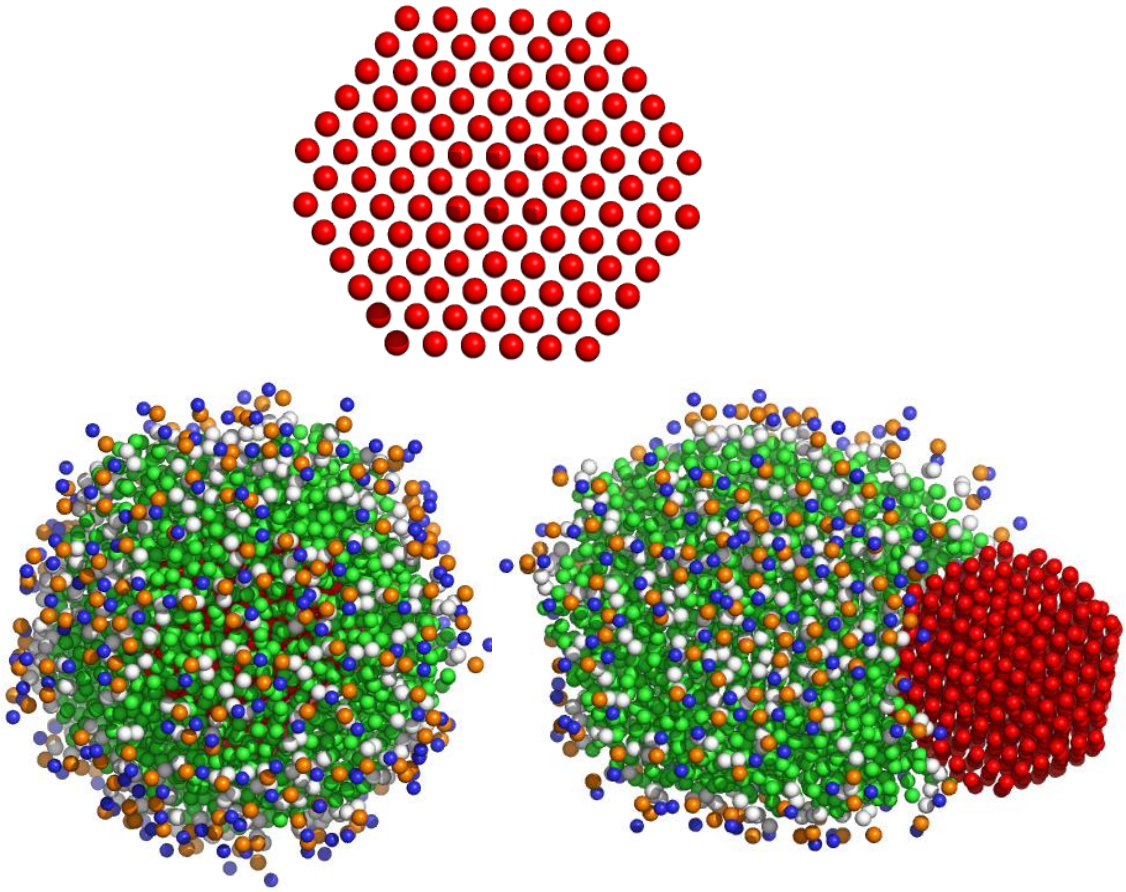


Figure S4: Snapshot of (*Upper panel*) a faceted NP (*Lower left panel*) corona structure around a faceted C5 NP and (*Lower right panel*) hemi-micelle structure attached to the faceted C7 NP.

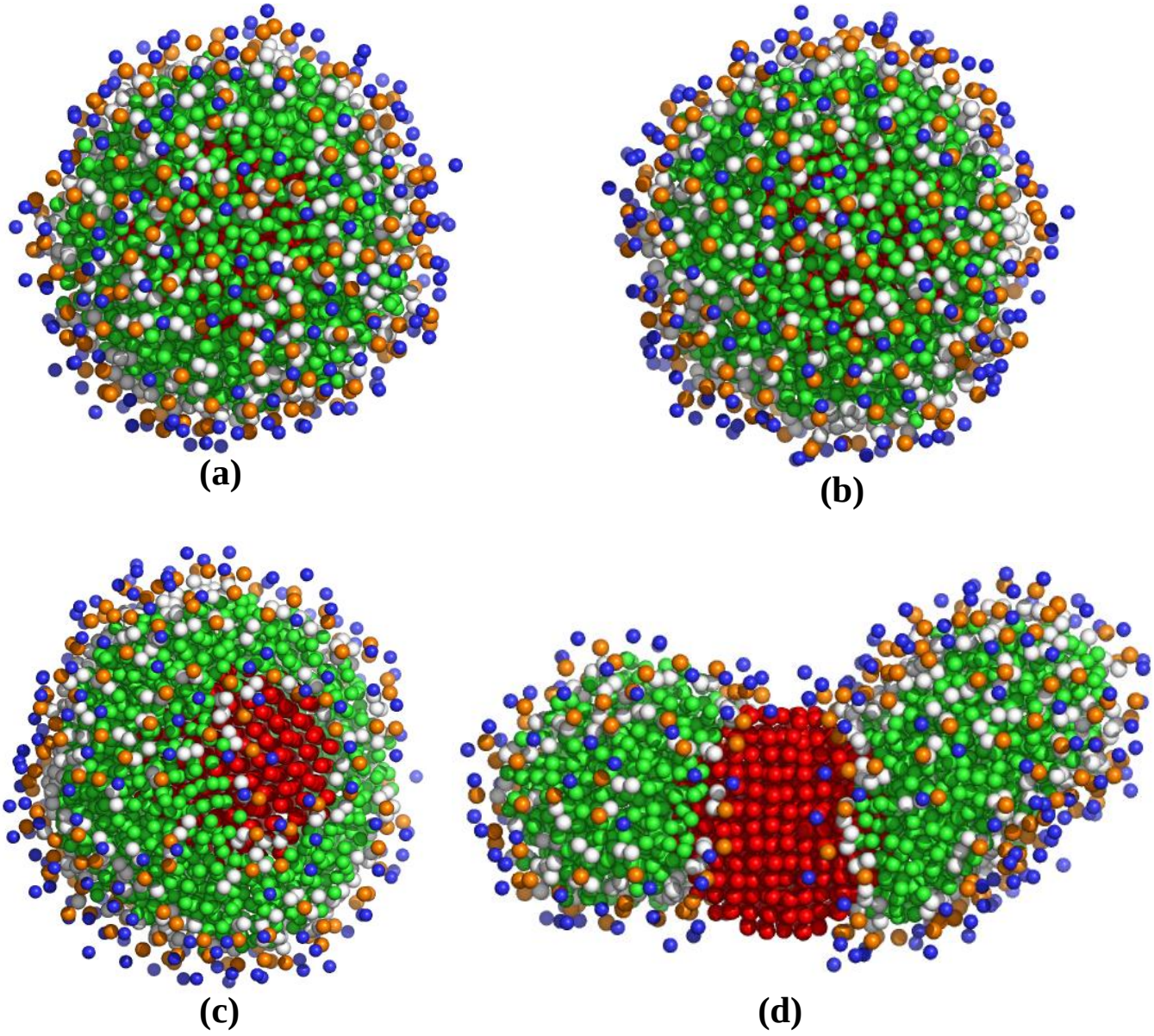


Figure S5: Corona structures of DPPC lipids around (a) C5, (b) N0, (c) Nda NPs. (d) hemi-micelles attached to the surface of a P1 NP. Structures are obtained from the BMW-MARTINI water model.

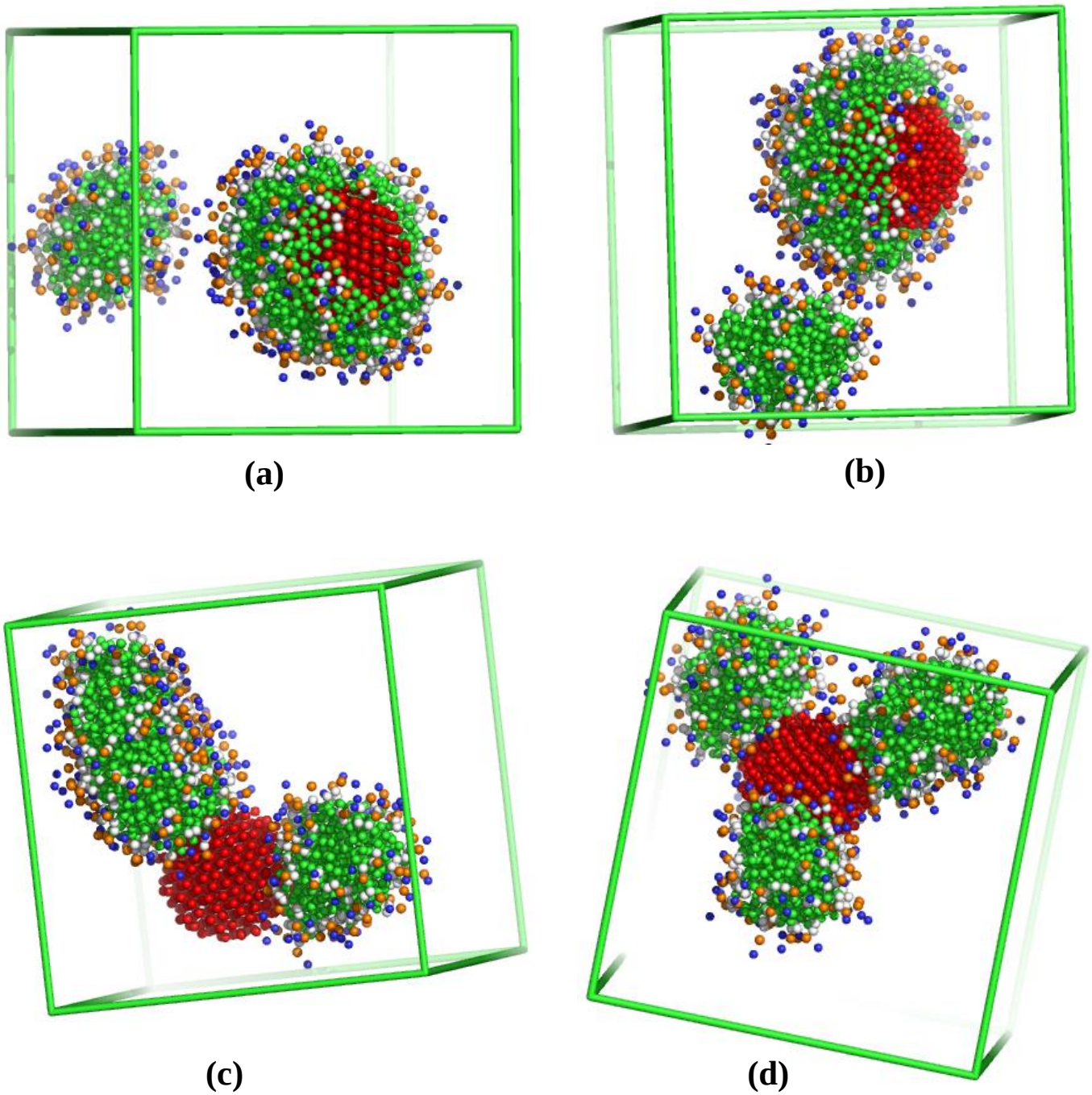


Figure S6: Corona structure of DPPC lipids around (a) C3 (b) C5 NP. Hemi-micelles attached to the surface of a (c) N0 and (d) Nda NP. Structures are obtained from the polarizable MARTINI water model.

Sequence of the SP-C peptide:

SER-PRO-PRO-ASP-TYR-SER-ALA-ALA-PRO-ARG-GLY-ARG-PHE-GLY-ILE-PRO-PHE-PHE-PRO-VAL-HIS-LEU-LYS-ARG-LEU-LEU-ILE-LEU-LEU-LEU-LEU

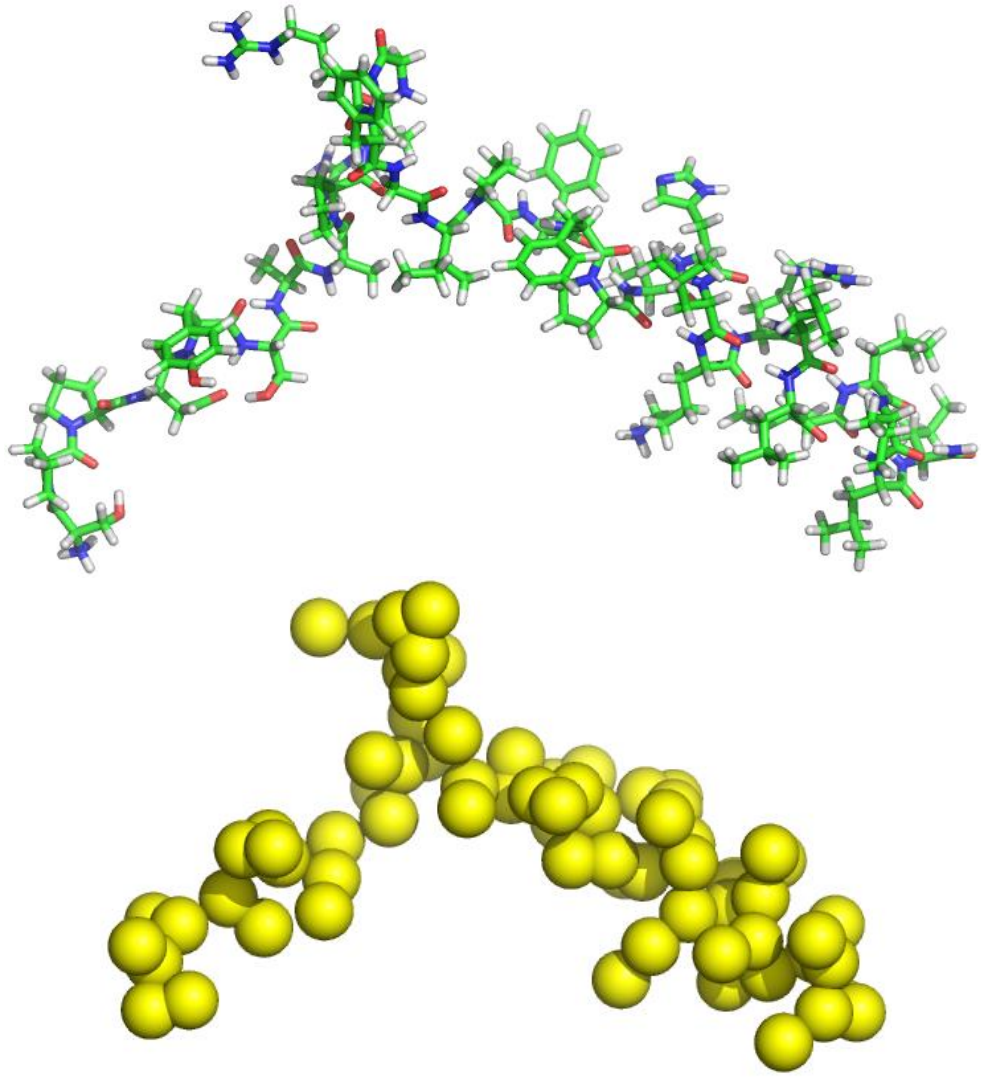


Figure S7: Atomistic (upper panel) and coarse-grained representation (lower panel) of the SP-C peptide. Green, blue, red and white colors represent carbon, nitrogen, oxygen and hydrogen atom, respectively. All of the CG beads are shown in yellow color. However, the mapping from atomistic structure to CG structure is done following the procedure as suggested in the MARTINI force field.

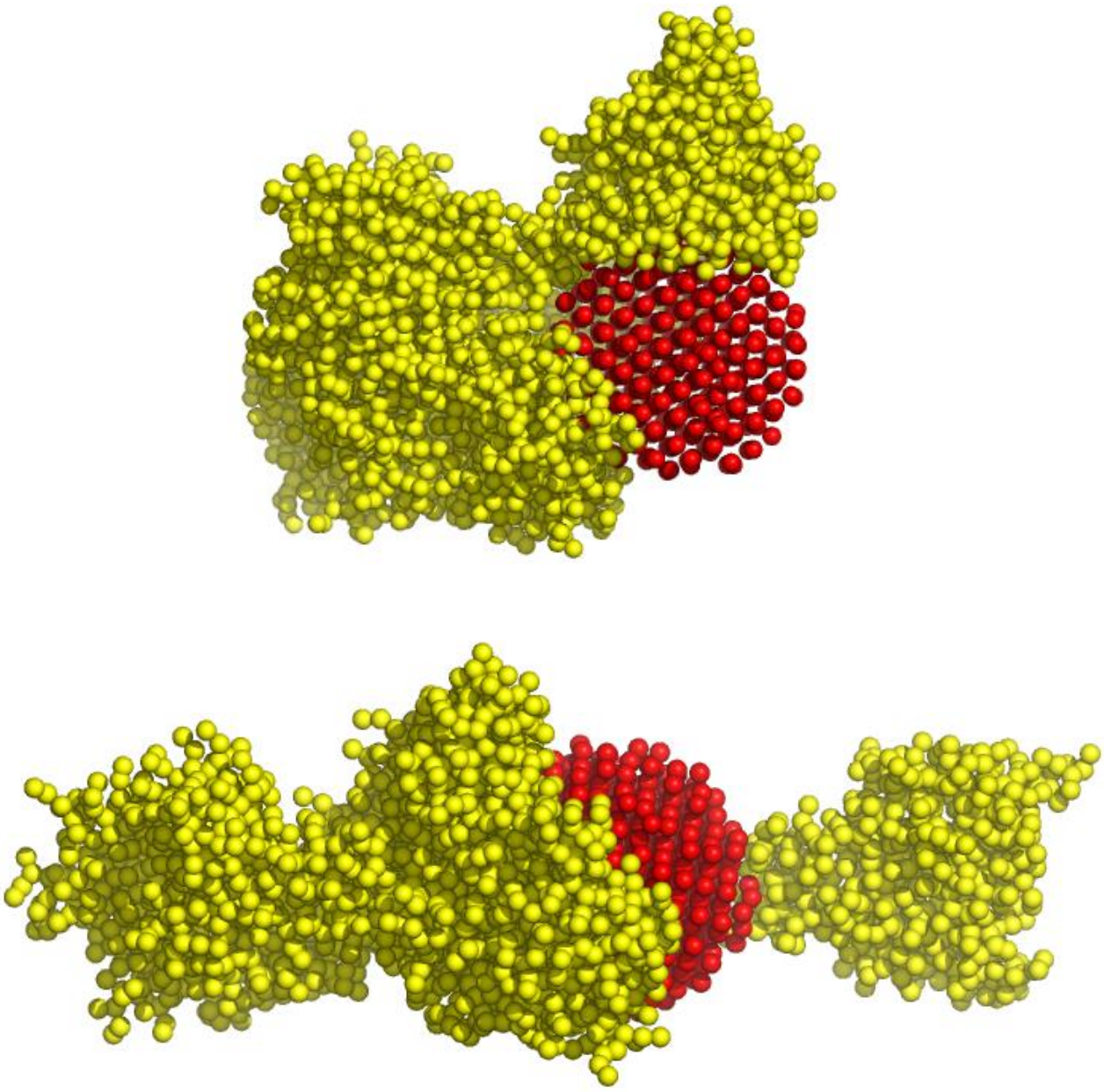


Figure S8: Snapshot of (*Upper panel*) C1-NP/SP-C peptide complex structure and (*Lower panel*) N0-NP/SP-C complex structure.

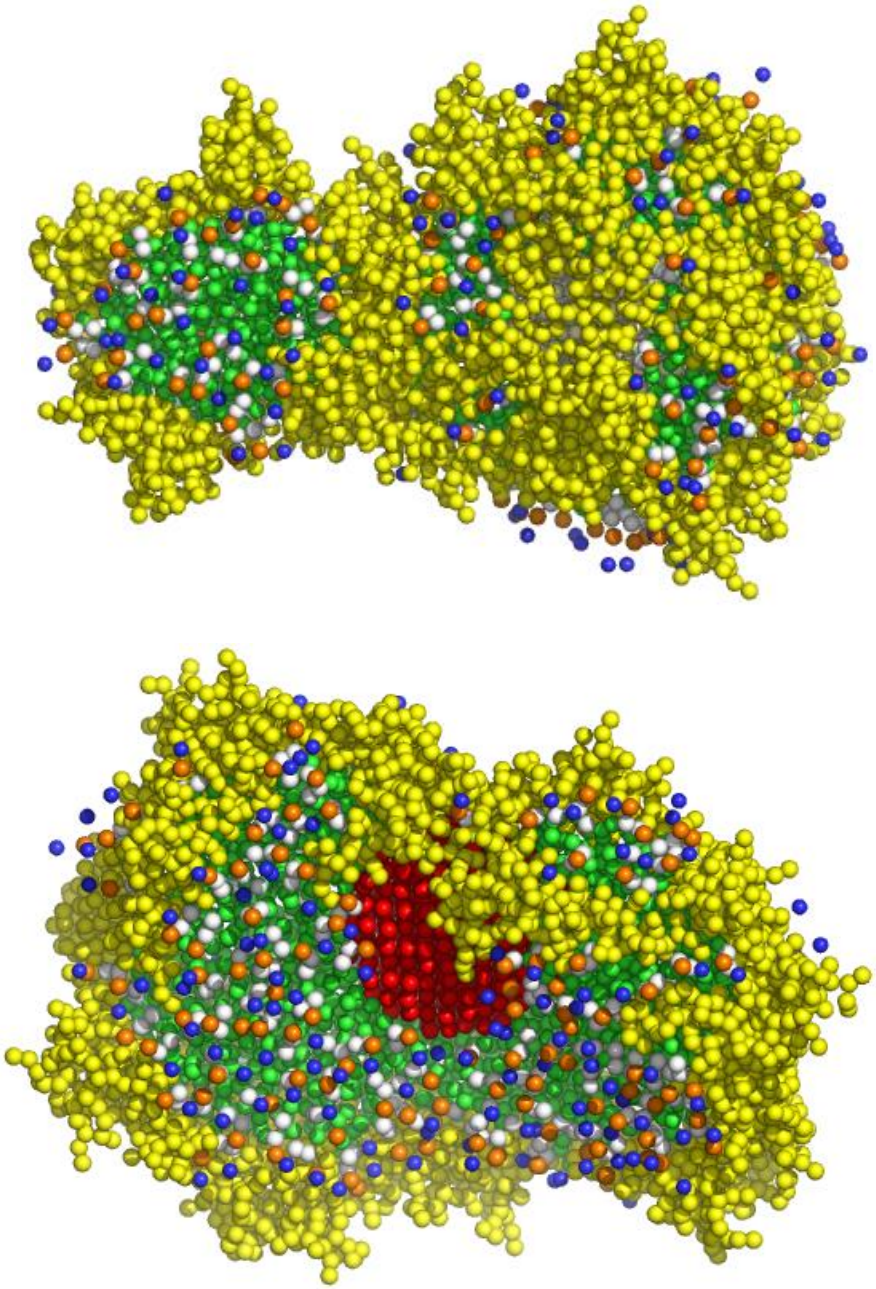


Figure S9: Snapshot of (*Upper panel*) C3-NP/DPPC/SP-C and (*Lower panel*) C5-NP/DPPC/SP-C complex structure in the presence of polarizable MARTINI water obtained after 1 μs-long simulation.