Electronic Supplementary Material (ESI) for Soft Matter.

Supporting Information

Figure S1 indicates the morphological evolution of the pure AB system as a function of a_{AW} and ϕ_{AB} (a_{AB} = 37.04, a_{BW} = 76.18). At a_{AW} = 20, AB polymers form small worm-like clusters, and the cluster number is increased by increasing concentration of AB (ϕ_{AB}), while the cluster size is nearly unaffected. At a_{AW} = 26.05, we obtain vesicle at $\phi_{AB} \leq 15\%$ and lamella at $\phi_{AB} \geq 20\%$. Specifically, at ϕ_{AB} = 20%, a complete lamella is obtained and additional polymers form local vesicles connected to the lamella; at ϕ_{AB} = 25% or 30%, an intersectional structure is obtained and again additional polymers form local vesicles connected to the structure (black regions indicate water clusters within or around the structure). At a_{AW} = 33, AB polymers form aggregated micelles with randomly distributed A and B blocks. The size of the micelle increases with increasing ϕ_{AB}, and we got sphere at $\phi_{AB} \leq 25\%$ and cylinder at ϕ_{AB} = 30%.

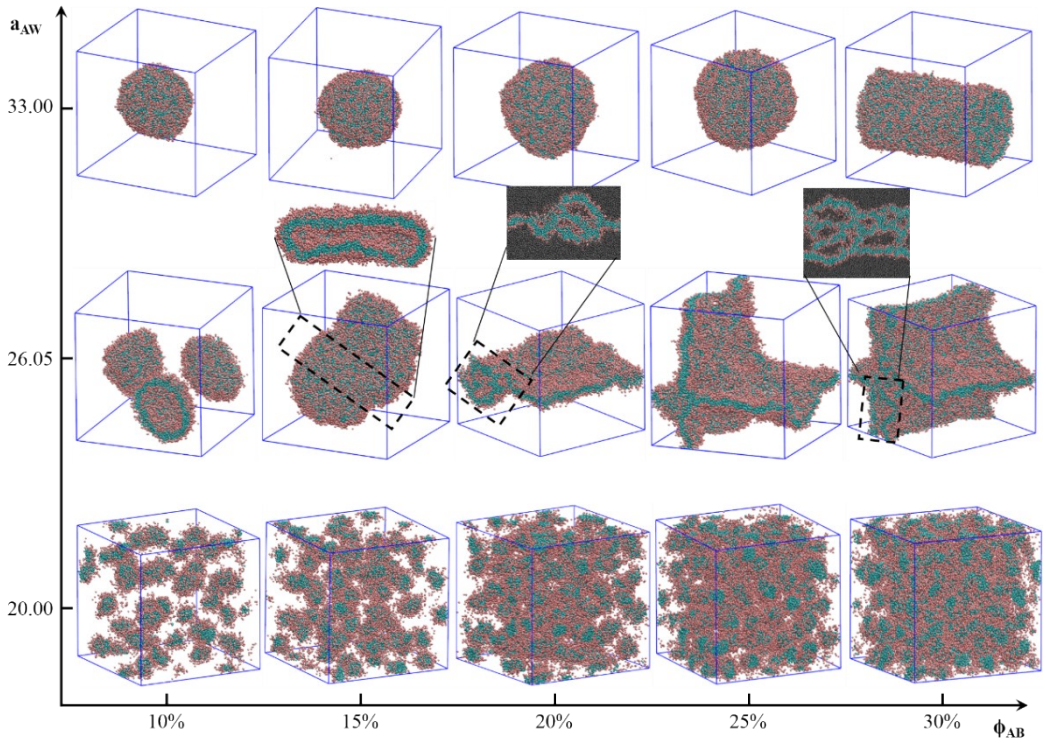

Figure S1. Morphologies of the AB system at various a_{AW} and ϕ_{AB} values (a_{AB} = 37.04, a_{BW} = 76.18).

Color scheme: pink-A, blue-B, black-water. Water is shown in the two local cross sections for better observation of the connected lamella-vesicle structure.

We further investigated the effect of box volume on the size of micelle. Compared with the micelles in Figure S1 at a_{AW} = 33 (box size: $45r_c \times 45r_c \times 45r_c$, bead number 2.73375×10^5), we increased the box size to $70r_c \times 70r_c \times 70r_c$ (bead number 1.029×10^6) and reran the simulation from a random state. The copolymers in both systems (ϕ_{AB} = 25% and 30%) gradually merge into a singular big micelle in the bigger box, while taking much longer time for the separated clusters to merge than that in the smaller box (Figure S2). Due to the computing limitation we have, we are not able to determine the threshold size of the micelle by further increasing the size of the box. In addition, we doubled the system size of the $45r_c \times 45r_c \times 45r_c$ system in two directions (i.e., $90r_c \times 90r_c \times 45r_c$, quadrupled volume). The micelles are complete stable at the continuation of the simulations (Figure S3). The dynamic motions of these micelles in this box can be seen in the attached video (*"motion_of_the_90rc-90rc-45rc_model.mp4"*), which indicates that the separated micelles basically keep stable (or fluctuate slightly) in the solution during the simulation. We also performed simulations with $(44r_c)^3$ and $(46r_c)^3$ system size keeping the particle number the same as in the $(45r_c)^3$ simulation where we again see stable simulations with slightly changed pressure again indicating that system size has only a minor effect (Figure S4). From above results, we believe that a well determined size distribution of the copolymer micelles can be obtained in a box that is larger than $(70r_c)^3$ which we cannot assess in our simulations. We presume that the reason why the copolymers have a large tendency to merge into a singular big micelle is due to the fact

that the block A we use is less hydrophilic (a_{AW} = 33). This increases the micelle/water interfacial tension, and makes it easier for block A to move inside the micelle (and form a multicompartment structure inside).

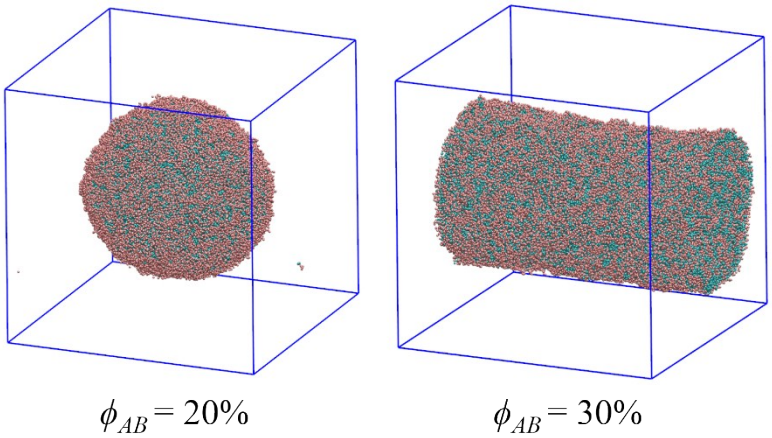

Figure S2. Morphologies of the AB system with box size of $70r_c \times 70r_c \times 70r_c$ at a_{AW} = 33 (a_{AB} = 37.04, a_{BW} = 76.18). Color scheme: pink-A, blue-B.

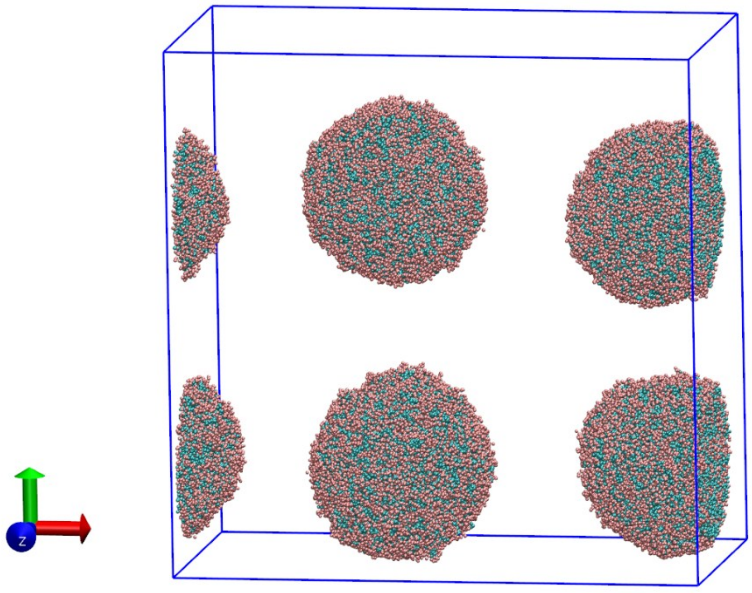

Figure S3. Morphology of the AB system with box size of $90r_c \times 90r_c \times 45r_c$ at a_{AW} = 33 (a_{AB} = 37.04, a_{BW} = 76.18). Color scheme: pink-A, blue-B.

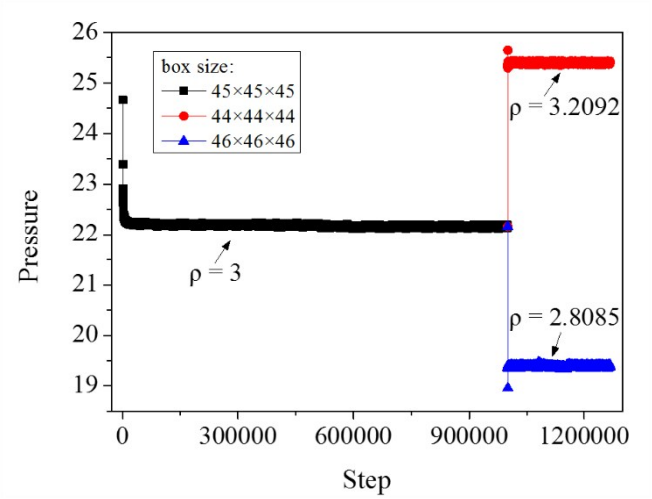

Figure S4. Pressure change of the AB system (ϕ_{AB} = 20%) when the box size $45r_c \times 45r_c \times 45r_c$ is slightly increased to $46r_c \times 46r_c \times 46r_c$ or decreased to $44r_c \times 44r_c \times 44r_c$ while keeping the bead number fixed.