

Electronic Supplementary Information

S1. Microaspiration

During this aspiration, optical microscope images of polymersomes are recorded on video and are analyzed to obtain a stretching modulus of the polymersomes and a critical tension where they rupture; stretching modulus (K_a) is a slope of tension (τ) over areal strain (α):

$$\tau = \frac{\Delta P \times R_p}{2 \left(1 - \frac{R_p}{R_s} \right)} \quad \text{and} \quad \alpha = \frac{\Delta A}{A_0} = \frac{2\pi R_p \Delta L}{A_0} \left(1 - \frac{R_p}{R_s} \right),$$

where ΔP is the suction pressure, ΔL is the length of deformed membrane in the pipette, A_0 is the original surface area of polymersome, and R_p and R_s are the radii of the pipette and the outer spherical contour of polymersome, respectively. (See Figure S2)

S2. Supplementary Figures

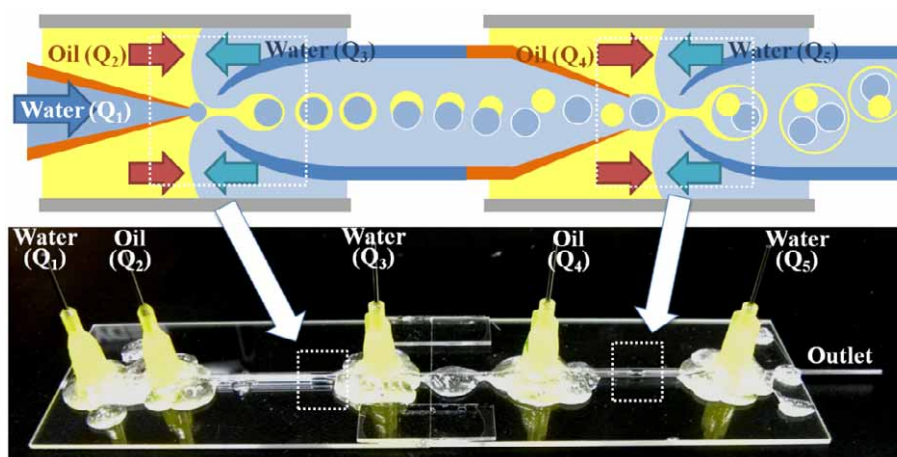


Fig. S1. Schematic illustration and image of the microfluidic capillary device with sequential double-emulsion drop-makers for preparation of W/O/W/O/W quadruple-emulsion drops.

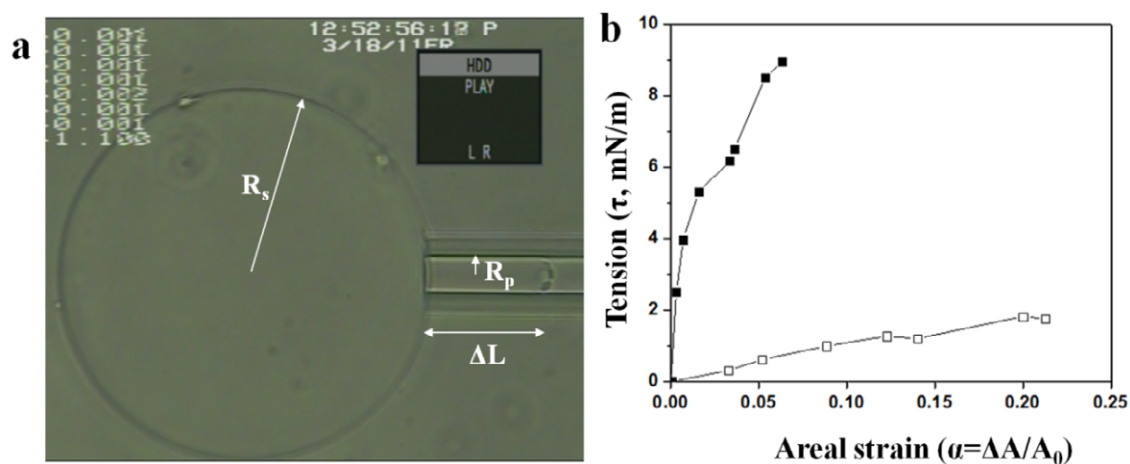


Fig. S2. (a) Parameters of aspiration experiment. (b) Areal strain-tension curves. The filled squares correspond to polymersomes without residual oil and the empty squares correspond to polymersome with residual oil, where the stretching modulus is slopes of these curves.

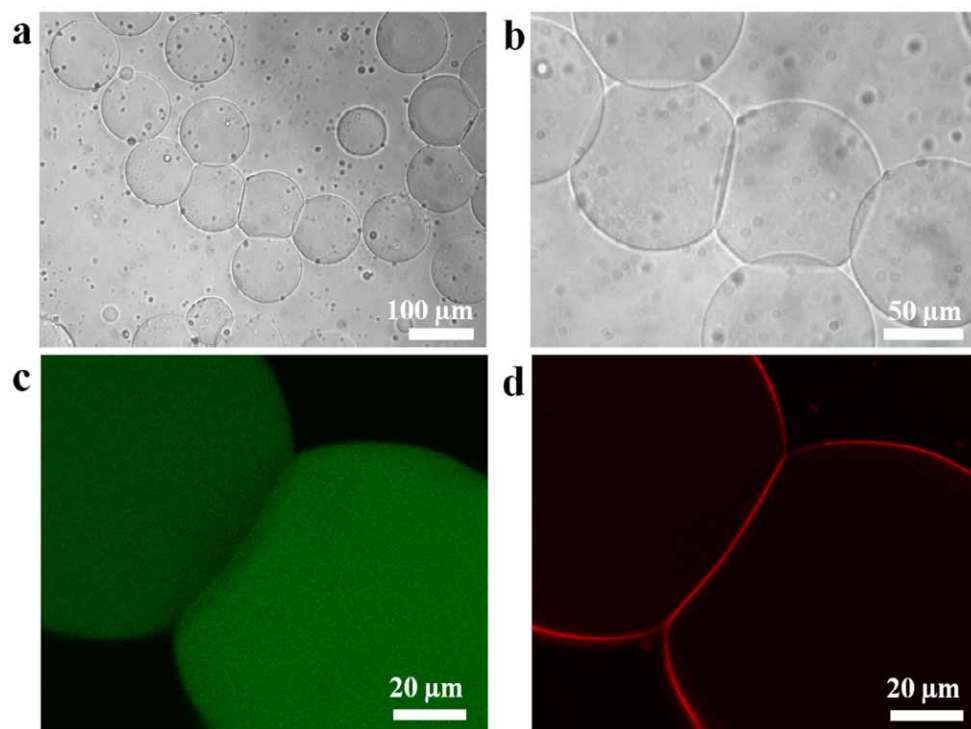


Fig. S3. (a, b) Optical and (c, d) confocal microscope images of polymersomes templated by double-emulsion drops; polymersomes are dispersed in a mixture of 25 mM of NaCl and 6% of PVA, where interior of polymersomes has green dye molecules of 8-hydroxyl-1,3,6-pyrenetrisulfonic acid, trisodium salt, while bilayer has red dye molecules of Nile red. The polymersomes form clusters by overlapping their bilayers.

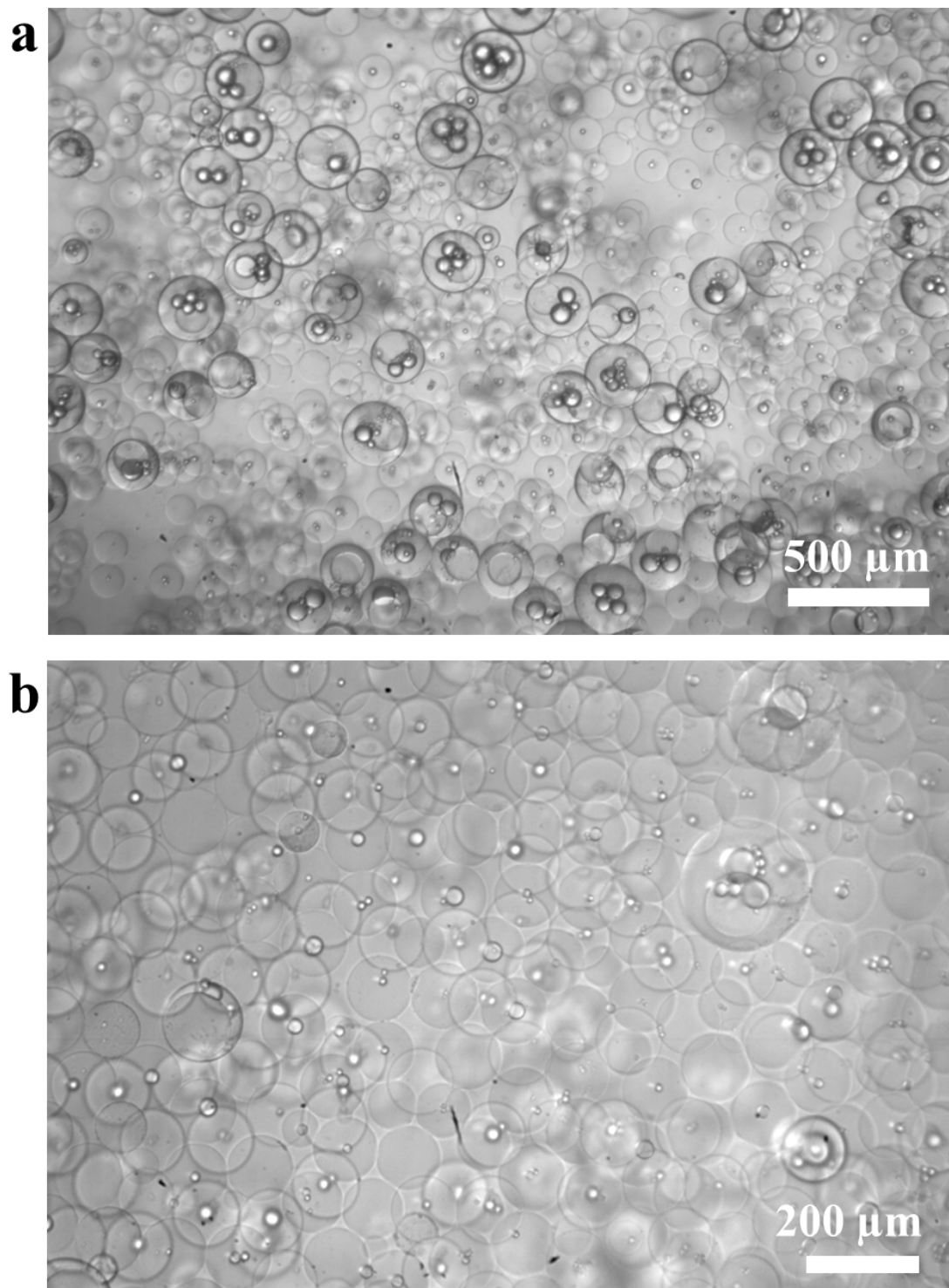


Fig. S4. Optical microscope images of polymersomes templated by quadruple-emulsion drops.

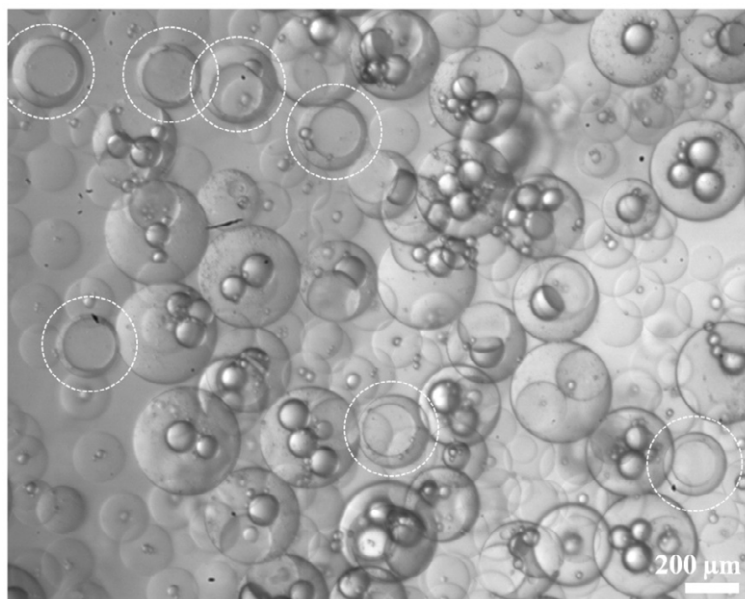


Fig. S5. Optical microscope image of polymersomes templated by quadruple-emulsion drops. When relative diameter of inner polymersomes to outer polymersomes is close to 1, a protrusion of the inner polymersomes is not dominant, resulting in polymersome-in-polymersome structure, as denoted by dotted circles.

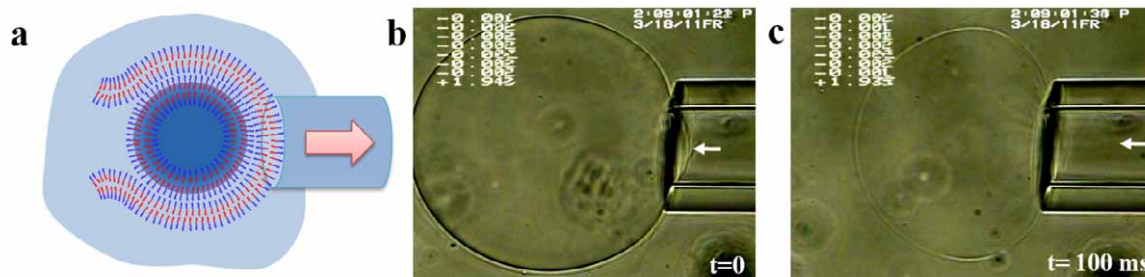


Fig. S6. (a) Schematic illustration of the micropipette aspiration experiment. (b, c) Optical microscope images of polymersomes (b) before and (c) after disintegration of the outer bilayer, where both the inner and the outer bilayers include PLA homopolymer in their hydrophobic regions.

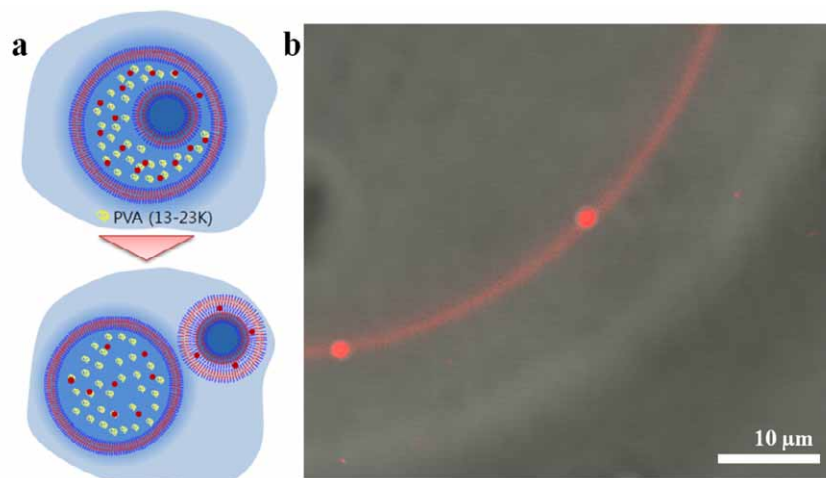


Fig. S7. (a) Schematic illustration of budding of the inner polymersomes, where the middle water phase contains 1 μm polystyrene particles. (b) Confocal microscope image of the polymersome produced by budding, which exhibits trapped polystyrene particles in its membrane.