Supplementary information:

Calculation of bubble size (R) and shell thickness (h):

The bubble radius (R) and shell thickness (h) are determined by mass balance using oil flow rate (Q_m), compound-bubble generation frequency (f_{cb}), volume fraction of polymer in oil (ϕ_p), and compound-bubble size (D_{cb}) with the images of compound bubbles generated in a device, as following:

The encapsulated bubble diameter (D_b) is calculated by $D_b = \left(D_{cb}{}^3 - 6Q_m / f_{cb}\pi\right)^{1/3}$. Then, the volume of polymer shell (V_p) upon the removal of solvent is calculated by $V_p = \phi_p V_o = \phi_p\left(V_{cb} - V_b\right) = \phi_p \frac{\pi}{6}\left(D_{cb}^3 - D_b^3\right)$, where V_o, V_{cb}, and V_b represent the volumes of the oil, compound bubble, and encapsulated bubble, respectively. Bubble radius (R) can be obtained using: $R = \frac{3}{4\pi}\left(V_p + V_b\right)^{1/3}$ and shell thickness (h) using: $h = R - D_b/2$.

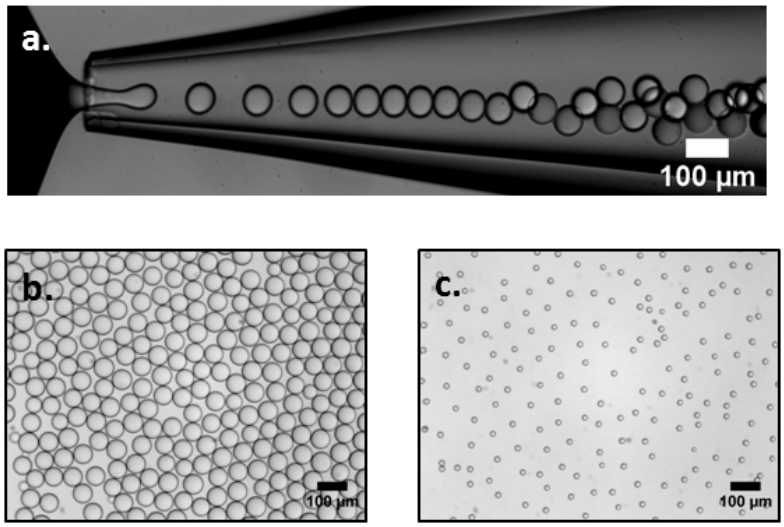

Figure S1. (a) Optical microscopy image of droplets of polymer solution generated in a flow-focusing microfluidic device. The inner oil phase is a 2 wt% PLGA solution in dichloromethane, while the outer phase is a 2 wt% PVA aqueous solution. (b,c) Optical microscopy images of polymer-oil droplets in water at (b) 90 seconds and (c) 780 seconds after preparation.

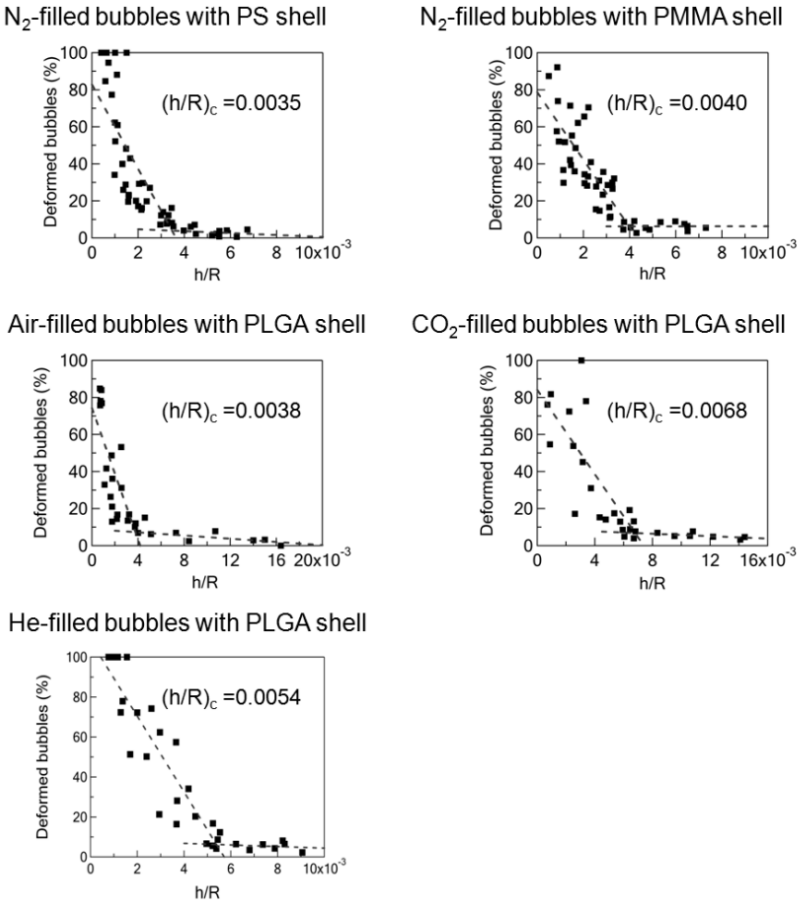

Figure S2. Percentage of deformed bubbles for different shell materials and different filling gases as a function of the *h/R* determined 30 minutes after preparation.

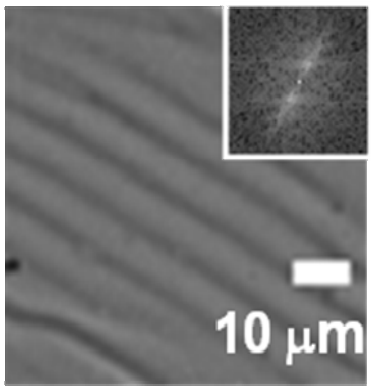

Figure S3. The optical microscopy image of buckled PLGA film with thickness of 225 nm on elastomeric PDMS substrate immersed in water for 30 minutes. Inset shows the fast Fourier transform (FFT) of the optical microscopy image.

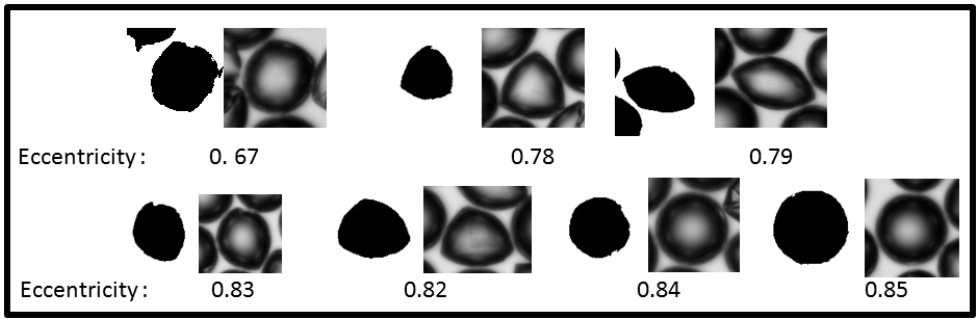

Figure S4. Restored and optical microscopy images of bubble-shape based on the value of eccentricity calculated using the ImageJ software. Eccentricity is defined as eccentricity = 4π(area/perimeter^2). Eccentricity values of 1.0 and 0.0 indicate a perfect circle and a line, respectively.