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Supplementary information for

Alginate Microgels Created by Selective Coalescence be-tween Core Drops Paired by Ultrathin Shell

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S1. Spontaneous separation of dual-cored double-emulsion drops from other drops

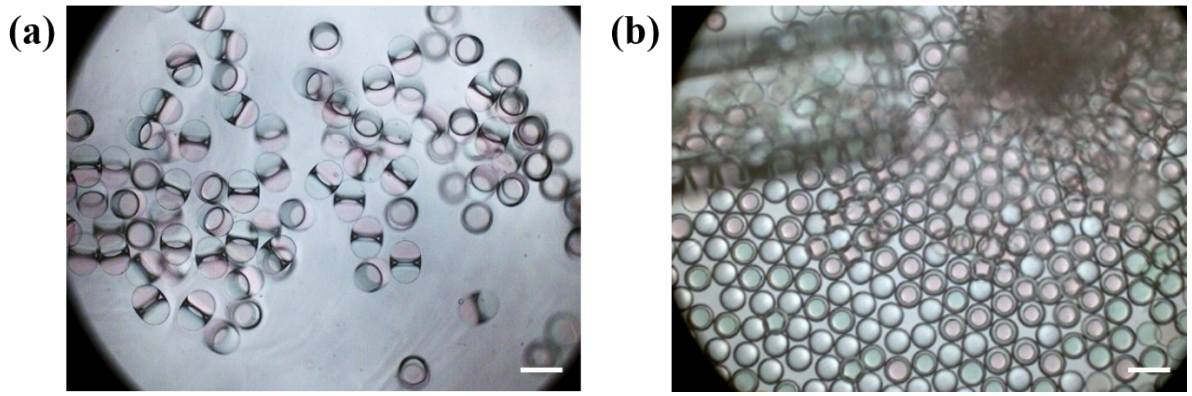

Figure S1. (a, b) Optical microscope (OM) images taken at collection bath with two different focal planes of (a) the bottom of the bath and (b) immediate underneath of air-collection liquid interface. All dual-cored double-emulsion drops sediment, while single-cored double-emulsion drops and single oil drops float. This is because density of collection liquid is set to be smaller than that of dual-cored double-emulsion drops with ultrathin shell and larger than those of other drops with large oil volume at the same time.

S2. Importance of two separate steps of drop formation and gelation

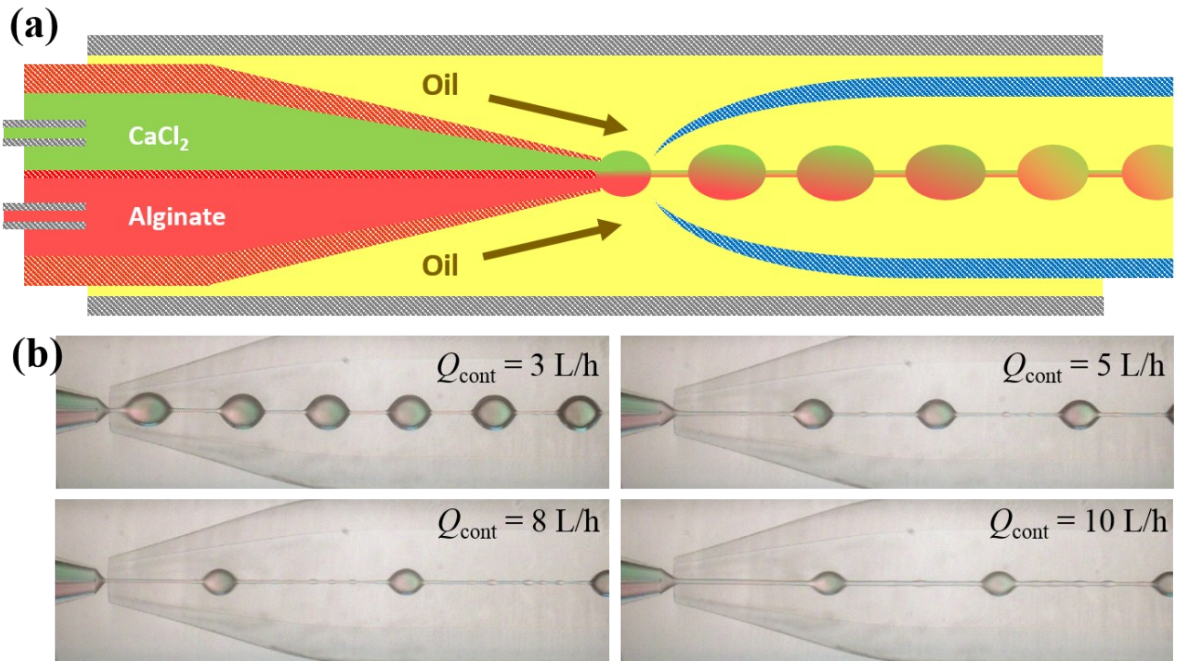

Figure S2. (a, b) Schematic illustration and OM images of a capillary microfluidic device showing the formation of gel drops connected by a thread, where alginate solution and calcium ion solution are simultaneously injected through two channels of theta-shaped capillary respectively and mineral oil is injected through interstices between the capillary and outer square capillary. Calcium ions rapidly gelate alginate molecules as soon as two solutions are brought into contact at the tip of theta-shaped capillary, forming viscoelastic thread. This prevents the formation of separate microgels even with high flow rate of continuous phase; flow rate of continuous phase is denoted in each panel of (b), where flow rate of two solutions is maintained at 300 μLh^{-1}.

S3. Size control of dual-cored double-emulsion drops.

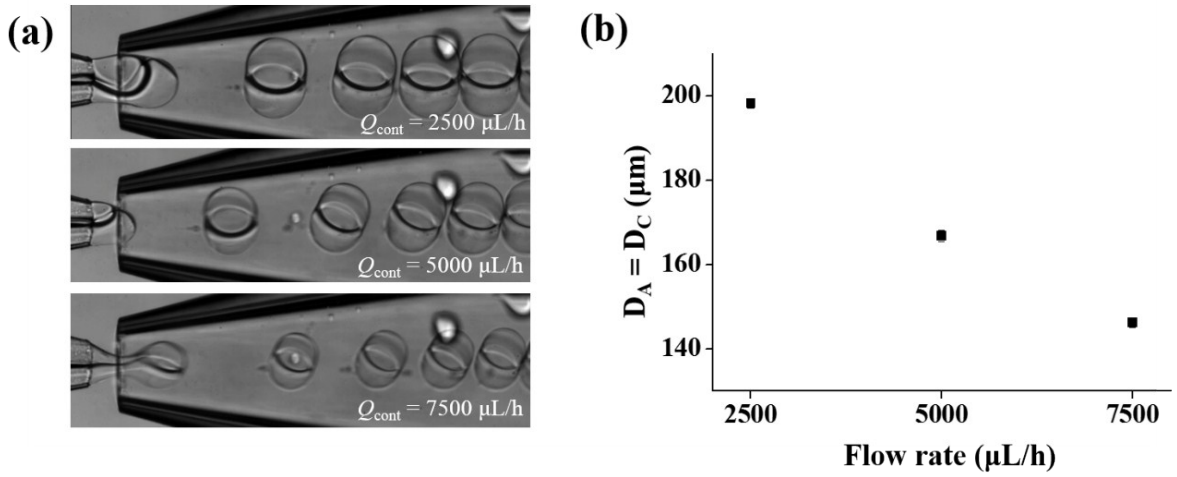

Figure S3. (a, b) OM images showing generation of monodisperse dual-cored double-emulsion drops with three different sizes, where flow rate of continuous phase is set to be 2500, 5000, 7500 µLh-1 from the top. (b) Influence of the flow rate of continuous phase on the diameter of core drops.

S4. Viabilities of encapsulated and free-living cells in unfavorable light condition

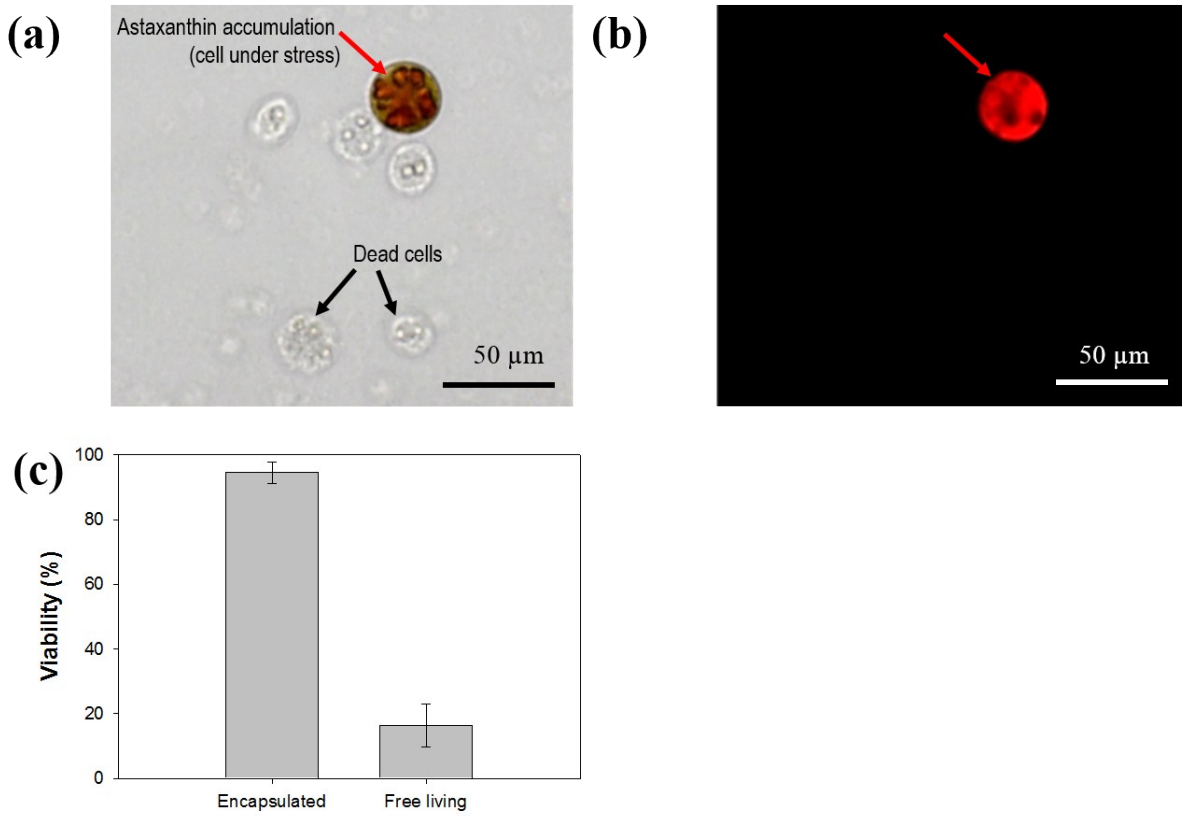

Figure S4. (a, b) Bright field and fluorescence microscope images of free-living H. pluvialis which is illuminated by white light with high intensity of 80 µmol $m^{-2}s^{-1}$ for 16 days. Under such an unfavorable condition, 83 % of free-living cells dies or enters resting stage as indicated with arrows, where cells at the resting stage turn red by accumulating astaxanthin. (c) Viability comparison between cells encapsulated in microgel and cells suspended without encapsulation (free living).

S5. Description of Movies

- Movie S1 shows generation of dual-cored double-emulsion drops with ultrathin shell.
- Movie S2 shows selective coalescence between two cores and subsequent gelation of alginate at the first part and rapid transfer of microgel into fresh water medium by oil shell rupture at the second part.
- Movie S3 shows formation of asymmetric drops with controlled size ratio of two cores as value of Q_A/Q_C is adjusted from 1 to 8.