Supplementary Information

MICROFLUIDIC EMULSIFICATION AND SORTING ASSISTED PREPARATION OF MONODISPERSE CHITOSAN MICROPARTICLES

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1. Materials

Medium molecular weight chitosan (M.W. 40 kDa) and tripolyphosphate (TPP) were obtained from Sigma (Sigma Chemical Co., St. Louis, MO). Distilled water (DI water) was filtered by a 0.22 nm syringe filter (Millipore Inc., Clifton, NJ) before use in the preparation process. The continuous phase: sunflower seed oil (viscosity 55 mPas, density 0.869 g/mL) was purchased from Uni-President Enterprises Corp., Taiwan. All other reagents were commercially available and of the highest grade.

2. Experimental procedure

First, the fluids of the center and side inlet channels were set up with oil (with 1% span 80) and an aqueous sample (1% Chitosan in 1% acetic acid) solution, respectively. Second, the fluids were then injected into the microfluidic chip by syringe pumps (kd Scientific KDS230) programmed by PCs. Finally, the sorted droplets were dripped into the TPP solutions. After 20 min of hardening, TPP-chitosan microparticles are formed.

A microscope was used to observe the experimental results. The image and detection system consist of an optical microscope (TE2000U, Nikon, USA) and a digital camera (Evolution color VF, Nikon, USA). The diameter of 100 microspheres was measured to provide an average size. Results were expressed as the means ± standard deviation ($n = 3$). The statistical difference was assumed to be significant when $P < 0.05$ by the two-sided Student t-test.

3. Numerical simulation

In order to investigate the separation mechanism in more detail we employed the finite volume simulation software CFD-ACE+ (ESI CFD, Huntsville, USA) to study the motion of the droplets in fluid. The simulation was performed using structured meshes in a spray module that tracks the droplets through the calculation domain. Within the model, the tracking of the liquid droplets (discrete phase) is achieved by solving the governing momentum-conservation equations in a Lagrangian frame of reference. For transient sprays to trace droplets motion in the fluid, the spray equations are integrated for all the parcels over a period of the global time step. Sub-time steps are utilized if the internally calculated spray time step is smaller than the global time step. For a spherical droplet, the equation of motion for the droplet can be written as:

$$\frac{dv}{dt} = \frac{3}{4}\frac{C_D \mu R_e}{\rho d^2}(U - v) + g \qquad (1)$$

where $v = ui + vj + wk$ is the velocity; u, v and w are the Cartesian velocity components; C_D is the drag coefficient; ρ, μ, and U are the density, dynamic viscosity and velocity of the surrounding fluid, respectively; d is the droplet diameter and g is the gravity vector. The equation accounts for the acceleration/deceleration of the droplet due to the combined effects of drag with fluid and body forces such as gravity. Under the condition of droplets motion in the microchannel, it is reasonable to neglect the gravity term in the analysis.

The simulation was performed using structured meshes with a total of 2088 nodes and 1860 cells. Furthermore, a transient simulation was performed in an explicit formulation using a first order Euler scheme with fixed time steps. At the same time, the model assumed a constant oil flow rate of inlet, and outlet boundary conditions were also defined by fixed velocities. The no-slip boundary condition was used on all other surfaces. For the sake of simplicity the simulation was conducted on a 2-D model. Densities of the water/oil flows are 1.000 and 0.869 g/mL, while the viscosities are 100 and 55 mPas, respectively. The water/oil fluids are assumed to be incompressible fluids and their physical properties are invariable.

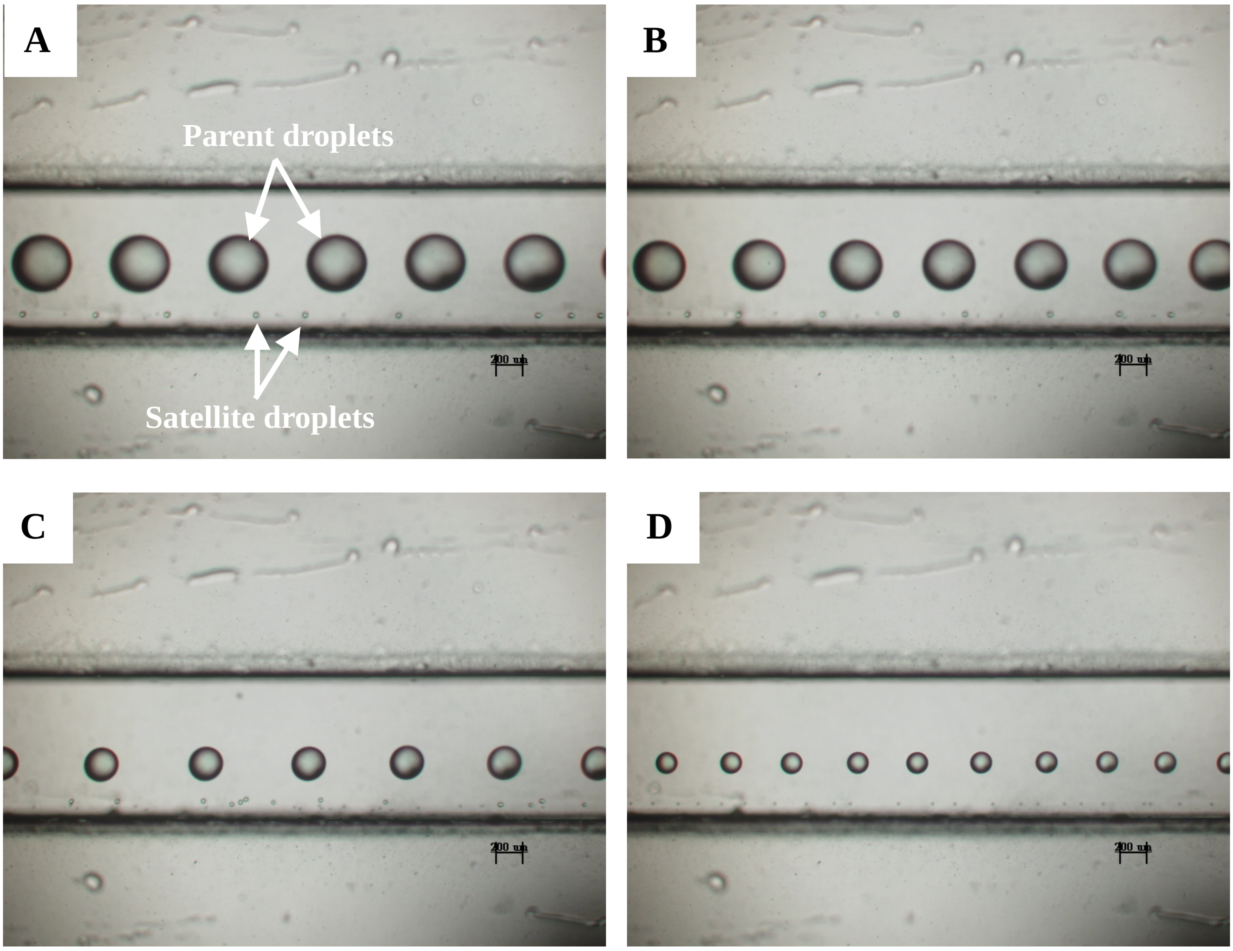

Figure S1 Photo images of the parent/satellite droplets generation in the microchannel. (Scale bar: 200 μm)

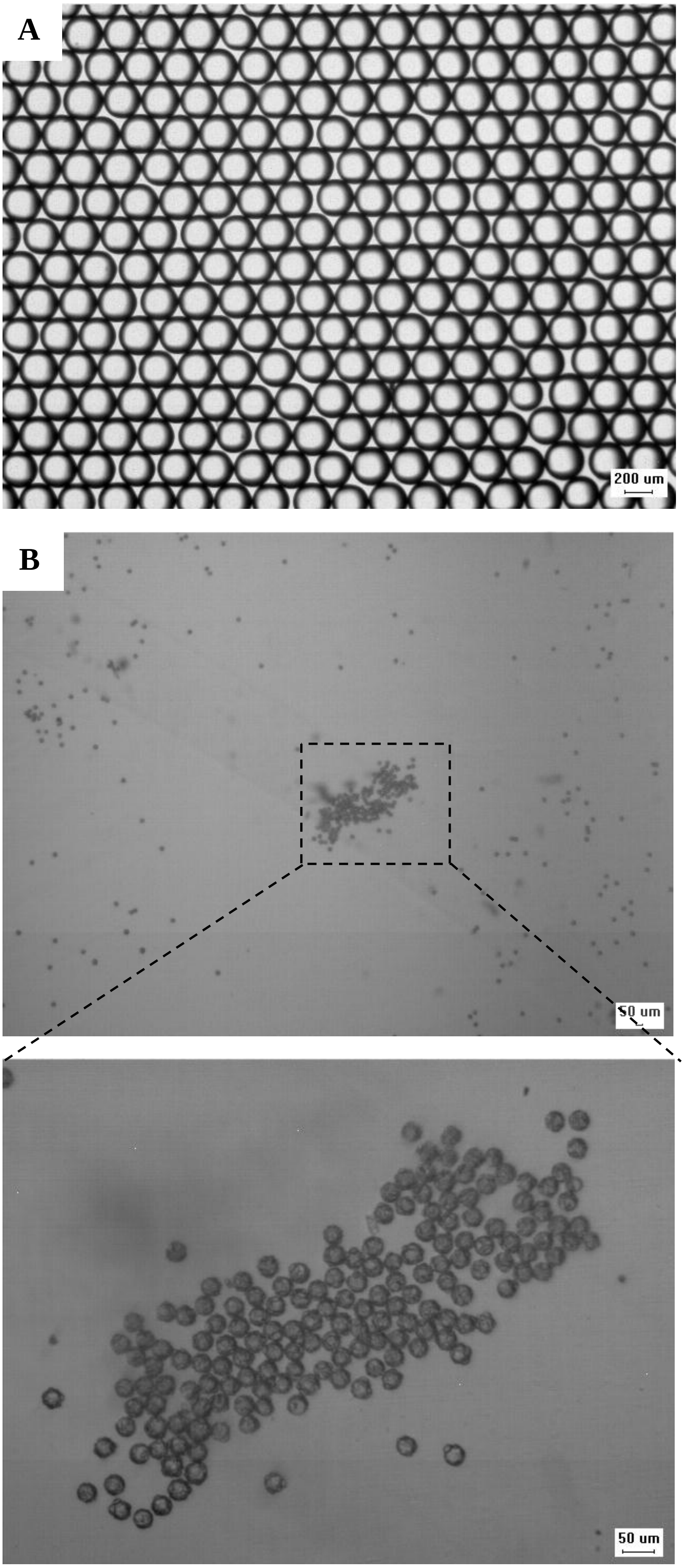

Figure S2 Optical microscopy image of the monodispersed chitosan microbeads (A) before and (B) after gelation.