Electronic Supplementary Information for

Producing Shape-Engineered Alginate Particles Using Viscoplastic Fluids

Sima Asadi,[a] Arif Z. Nelson,[b‡] and Patrick S. Doyle*[abc]

[a] *Department of Chemical Engineering, Massachusetts Institute of Technology, Cambridge, MA 02139, USA. E-mail: pdoyle@mit.edu*

[b] *Critical Analytics for Manufacturing Personalized-Medicine, Singapore-MIT Alliance for Research and Technology, Singapore 138602, Singapore.*

[c] *Harvard Medical School Initiative for RNA Medicine, Boston, MA 02215, USA.*

‡ *Present address: Food, Chemical and Biotechnology Cluster, Singapore Institute of Technology, Singapore 138683, Singapore.*

This pdf file includes Figure S1, Figure S2, Figure S3, and the captions for Movies S1 to S4. Movies S1 to S4 have been provided separately as mp4 files.

Droplet diameter calculation:

We used 4 different methods to calculate the diameter of viscoplastic droplets dripped from a 14G vertical nozzle:

1. Dripping time of $n = 10$ droplets, t, was used to calculate $D_M = (6Q_0 t/(n\pi))^{1/3}$, where Q_0 is the fluid flow rate set by a syringe pump.

2. Projected area of the droplet obtained from its 2D image, A, was used to calculate $D_A = (4A/\pi)^{1/2}$.

3. Volume of the droplet was used to calculate $D_V = (6V/\pi)^{1/3}$. Assuming rotational symmetry, we calculated $V = \int_0^L \pi x^2 \, dy$, where L and $x = f(y)$ are the droplet length and radius, respectively. $f(y)$ was obtained by fitting a 6^{th} degree polynomial curve to the semi-perimeter of the droplet in its 2D image.

4. Similar to method 3, volume of the droplet was used to calculate $D_C = (6V_C/\pi)^{1/3}$. We used the length of droplet centerline in the 2D image, L_C, and its distance from the droplet edge, $x_C = f(y)$, and calculated $V_C = \int_0^{L_C} \pi x_C^2 \, dy$.

The results of all methods have been shown in Fig. S1.

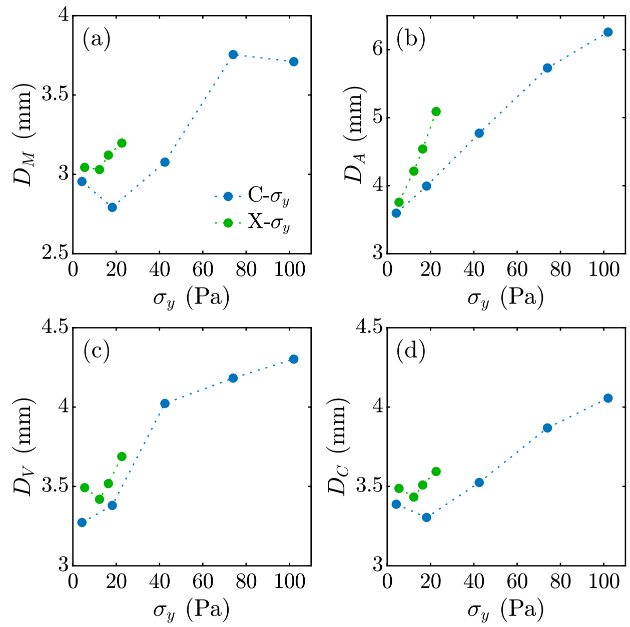

Figure S1: Diameter of C-σ_y and X-σ_y droplets dripped from a 14G vertical nozzle. (a) D_M, (b) D_A, (c) D_V, and (d) D_C have been defined above. Each data point in (b), (c), and (d) is the average of 3 trials and the error bars (not shown) are smaller than the marker size. Dotted lines are used as a guide for the eyes.

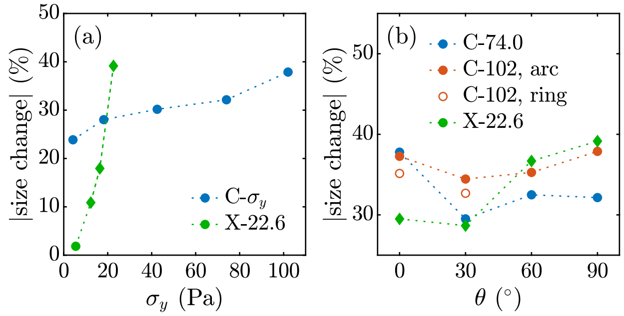

Figure S2: Effect of (a) σ_y and (b) θ on the degree of shrinkage and swelling of C-σ_y and X-σ_y particles compared to the droplets. Circle markers show negative values (shrinkage) and the diamond markers show positive values (swelling). Each data point is the average of 3 trials and the error bars (not shown) are smaller than the marker size. Dotted lines are used as a guide for the eyes.

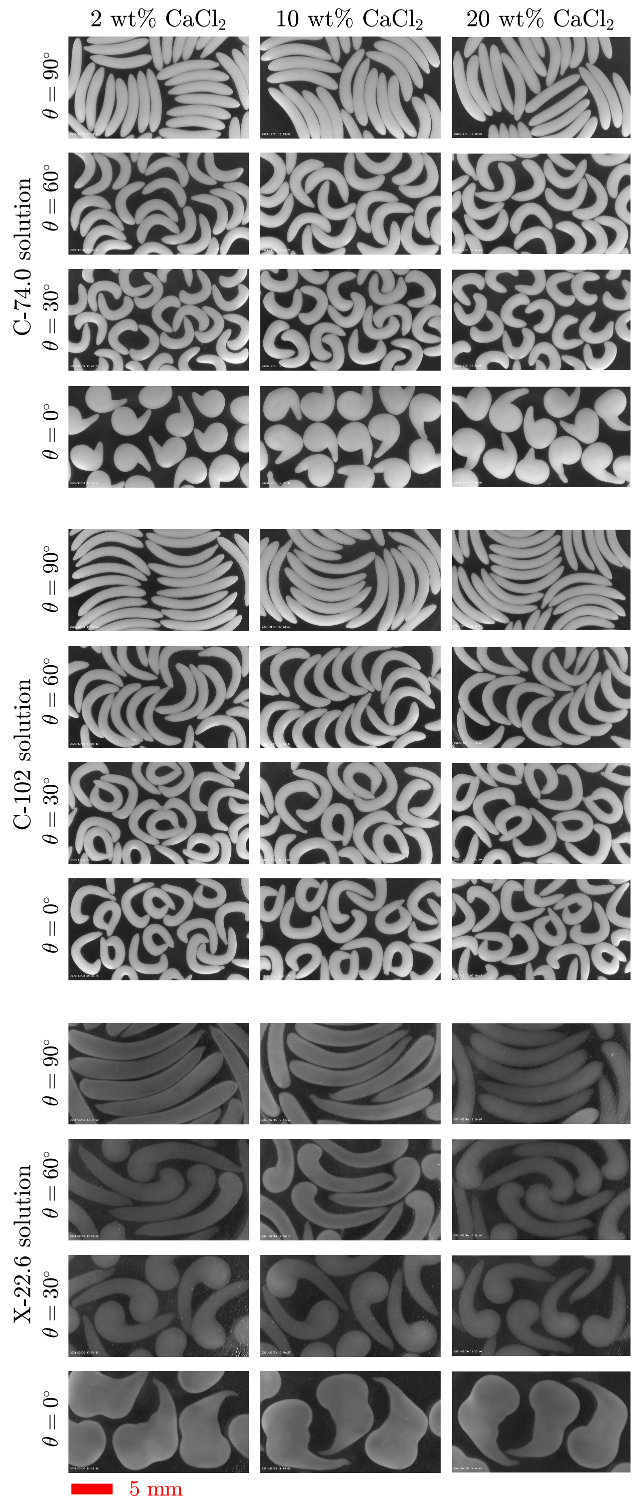

Figure S3: Effect of $CaCl_2$ concentration on the shape of alginate particles. Cumulative images of all particles produced using C-74.0, C-102, and X-22.6 solutions dripped from a 14G nozzle positioned at $\theta = 90°$, $\theta = 60°$, $\theta = 30°$, or $\theta = 0°$ into a 2, 10, or 20 wt% $CaCl_2$ bath. The red scale bar is 5 mm for all images.

Movie S1: C-σ_y solutions are dripped from a vertical 14G nozzle into a 2 wt% $CaCl_2$ bath. The red scale bar is 5 mm for all videos.

Movie S2: C-74.0 and C-102 solutions are dripped from a 14G nozzle angled at $\theta = 60°$, $\theta = 30°$, or $\theta = 0°$ into a 2 wt% $CaCl_2$ bath. The red scale bar is 5 mm for all videos.

Movie S3: X-σ_y solutions are dripped from a vertical 14G nozzle into a 2 wt% $CaCl_2$ bath. The red scale bar is 5 mm for all videos.

Movie S4. X-22.6 solution is dripped from a 14G nozzle angled at $\theta = 60°$, $\theta = 30°$, or $\theta = 0°$ into a 2 wt% $CaCl_2$ bath. The red scale bar is 5 mm for all videos.