Electronic Supplementary Material (ESI) for Reaction Chemistry & Engineering.


## Supplementary Figures

# Scalable preparation of macroporous collagen microgels by air bubble-induced breakup and ice templating

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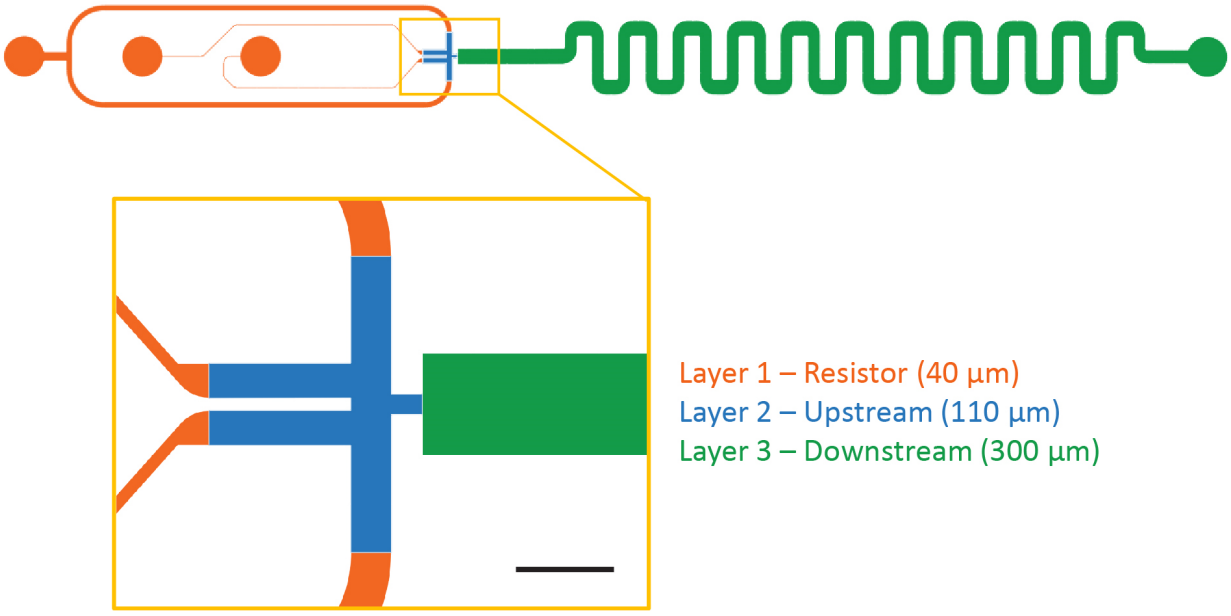


**Fig. S1** Schematic illustration of three different microchannel feature layers that are aligned to define a flow focusing generator (FFG) for air-bubble induced collagen droplet formation. The three layers correspond to different masking layers. Scale bar: 400 µm.

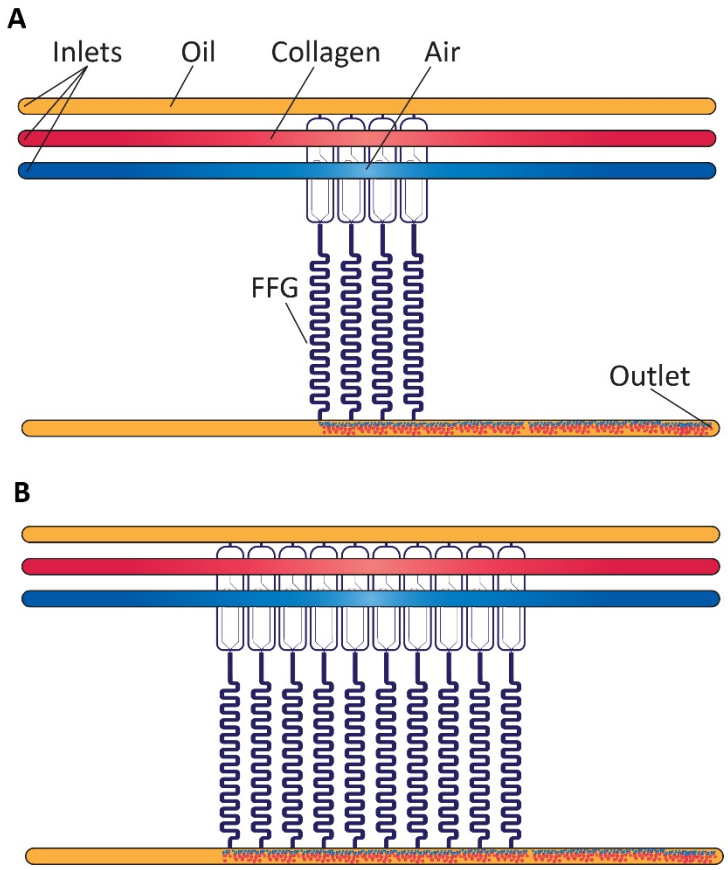


**Fig. S2** Device design for droplet generator with 4 FFGs and 10 FFGs (A) and (B), respectively.

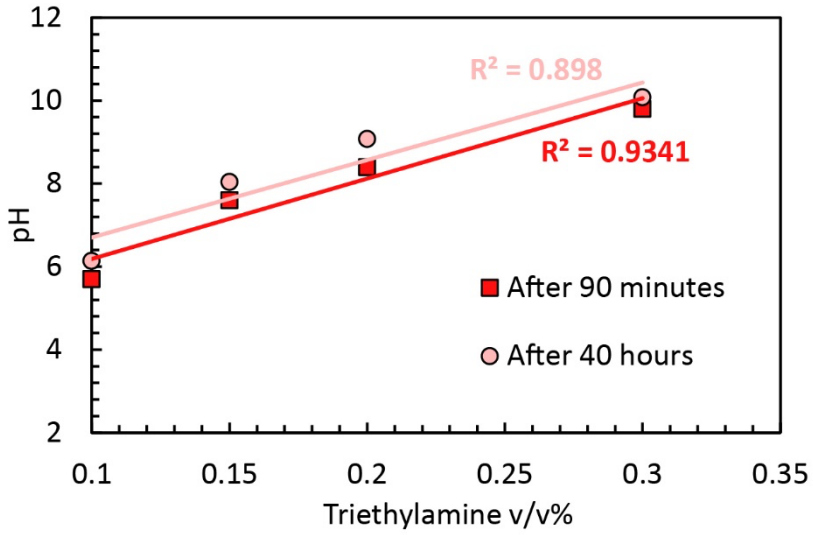


**Fig. S3** pH values obtained for different volume concentrations of triethylamine 1.5 h and 4 h after mixing.

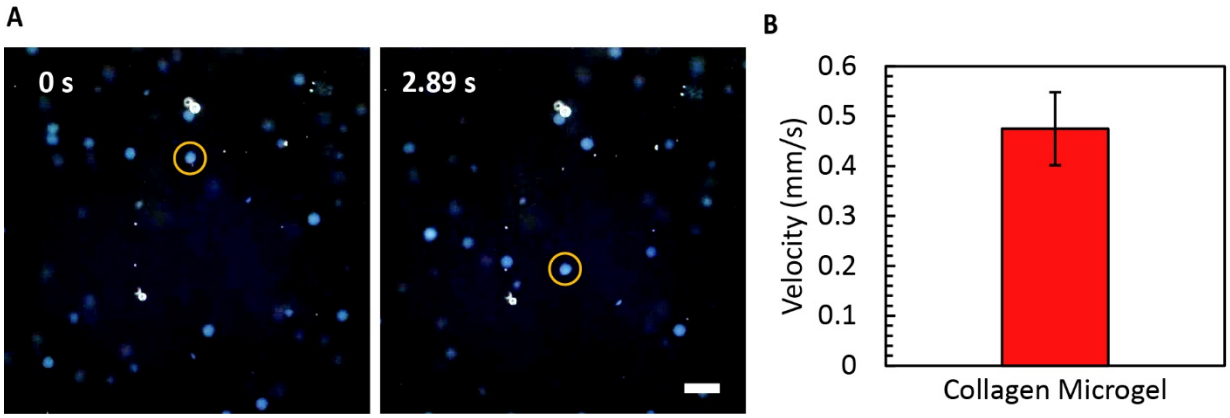


**Fig. S4** (A) Representative bright field micrograph obtained during the settling of collagen microgels in deionized water at room temperature. Scale bar: 500 µm. (B) Settling velocity obtained from (A).

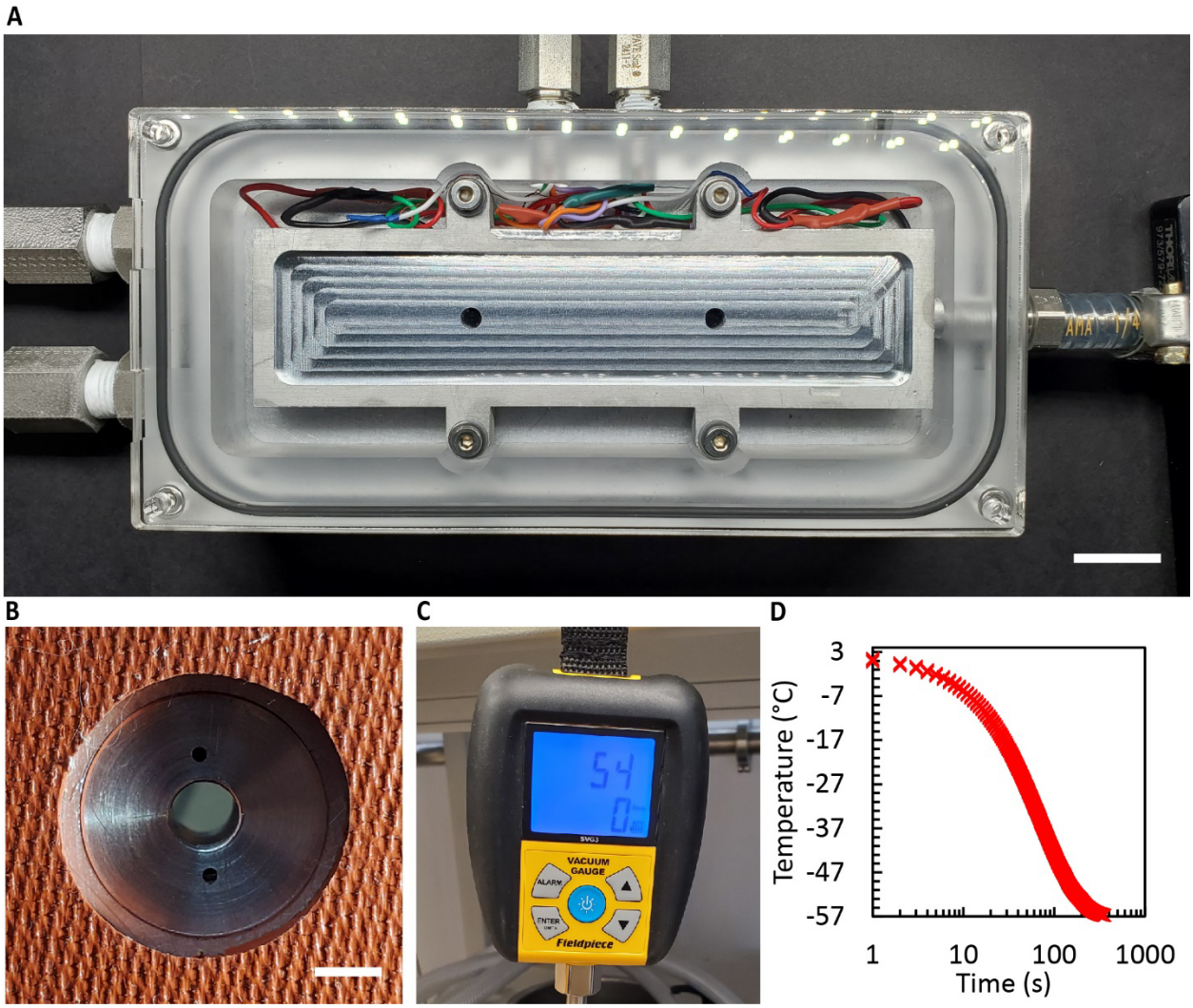


**Fig. S5** (A) Photograph of custom freezing setup developed for ice templating with a vacuum line attached on the right side. Fluidic connectors to an external chiller for coolant perfusion of the heat sink adjacent to the warm side of the TE elements (covered by sample chamber) are on the left. Scale bar: 2 cm. (B) Sapphire window for imaging MCM sample within the sample chamber. Scale bar: 0.5 cm. (C) Setup allows samples to be cooled to temperatures as low as -57°C and provides vacuum pressures as low as 7.2 Pa (54 mTorr). (D) Attainable chamber temperature without load (no MCM sample in the chamber). The triple point of water at 273.16 K is 612 Pa (4.6 Torr).

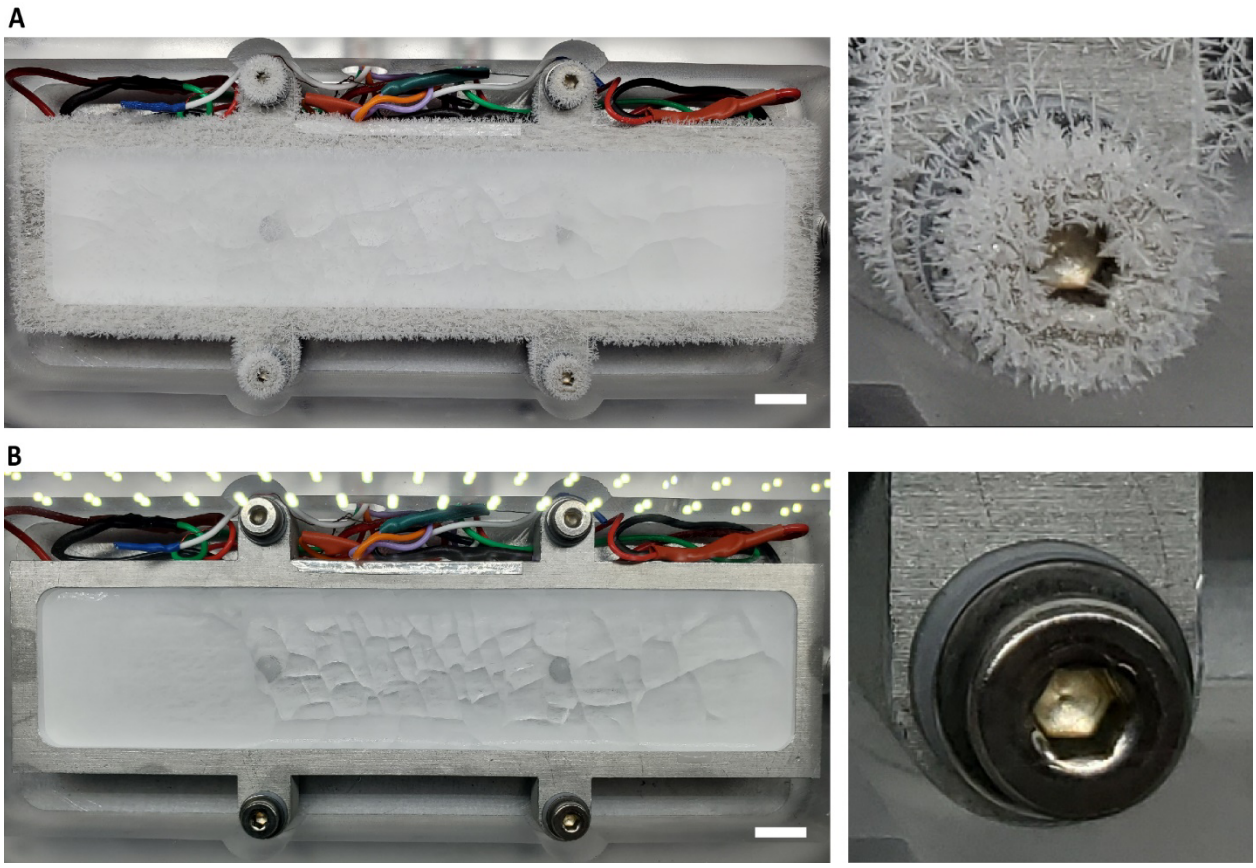


**Fig. S6** Ice templating without a lid in humid air causes water vapor to condensate and freeze, thereby increasing the thermal load on the system and reducing cooling performance. (B) When lid is attached and sealed and sample chamber purged with dry argon, no condensate is formed during freezing. Scale bar: 1 cm (A and B).


**Fig. S7** Samples can be lyophilized in situ within a sample chamber. (A) Frozen microgels in deionized water, ice templated at a cooling rate of 10°C/min (10 ml). (B) Freeze-dried samples after 24 h. Scale bar: 1 cm (A and B). (C) Freeze-dried sample after collection. Scale bar: 1 cm.

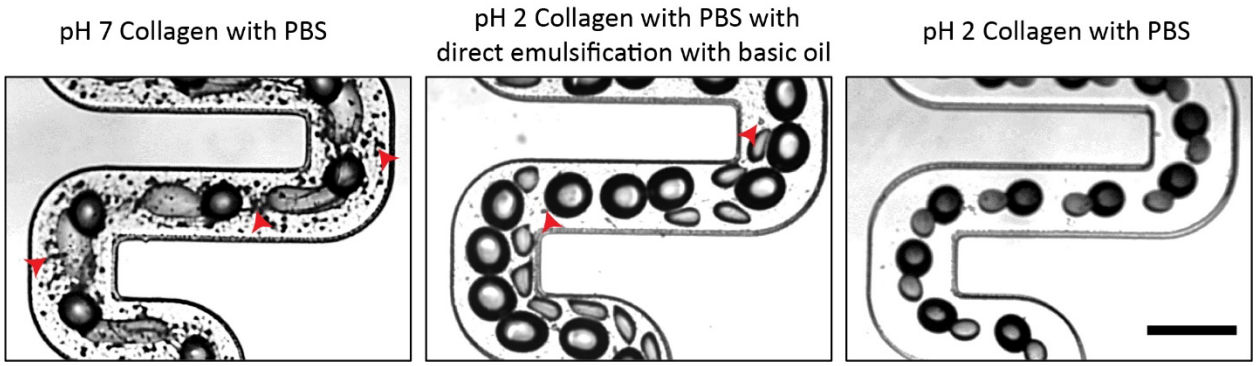


**Fig. S8** Red arrows point to collagen fragments at the three different conditions. Scale bar: 400 µm.

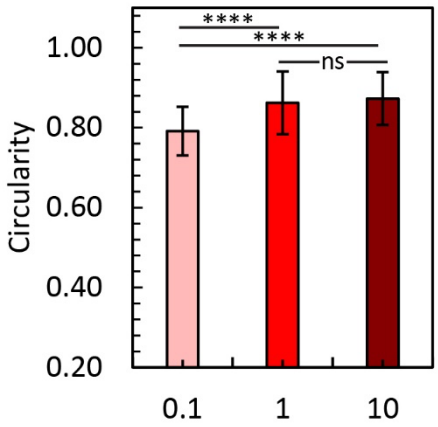


**Fig. S9** Circularity of MCMs after ice templating. Circularity $C = 12.57\frac{A}{P^2}$, A denotes microgel cross-sectional area and P perimeter. Ordinary one-way ANOVA with Tukey's multiple comparisons test was performed for statistical analysis. ns: Nonsignificant change.

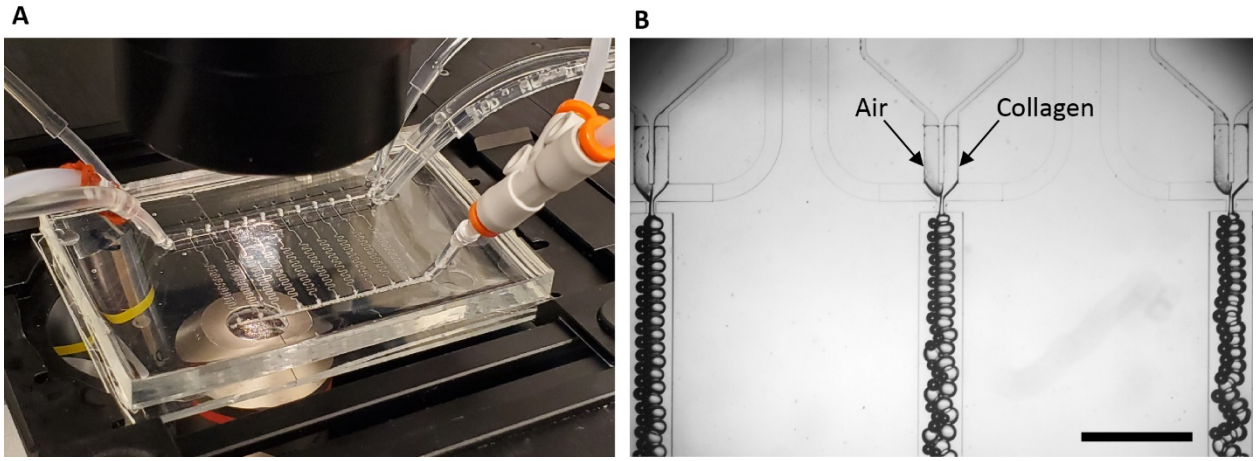


**Fig. S10** (A) Photograph of microfluidic device with 10 FFGs during continuous formation of collagen microgels using air-bubble induced breakup. (B) Closeup brightfield micrograph with three FFGs operating in parallel during air-bubble induced breakup of collagen microgels. Scale bar: 1 mm.

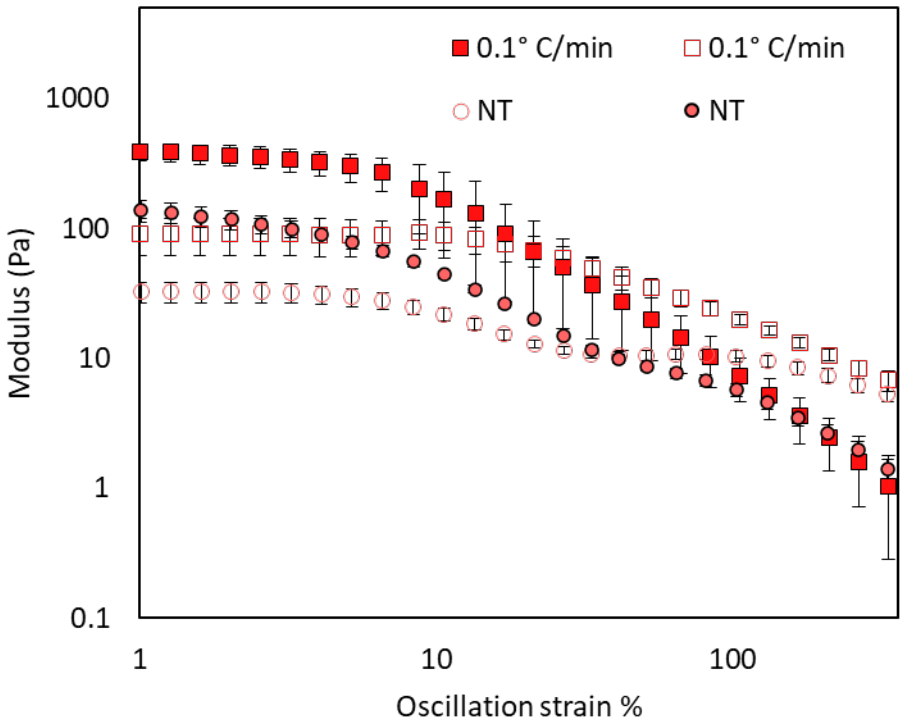


**Fig. S11** Rheological characterization of granular material prepared for non-ice templated and ice templated microgels (MCMs). Amplitude sweep data for ice templated and non templated (NT) microgels. Storage modulus (closed symbols) and loss modulus (open symbols) for same bioinks.