Electronic Supplementary Material (ESI) for Soft Matter.

SI Processing of fast-gelling hydrogel precursors in microfluidics by electrocoalescence of reactive species

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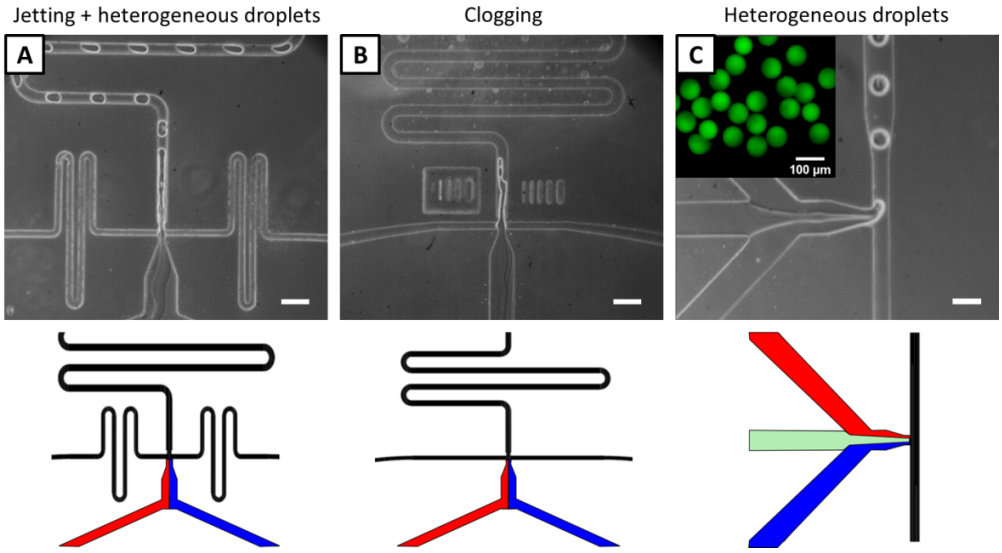

Figure S1: Phase-contrast microscopy images of common challenges in droplet microfluidics focusing on the processing of fast-gelling hydrogel precursors. (A) Jetting instead of geometry-controlled droplet formation at the microchannel cross-junction. (B) Clogging of the microfluidic droplet-forming junction due to in-channel hydrogelation. (C) Separation of two reactive aqueous precursor phases by a third, middle-phase flow acting as an unreactive spacer to prevent gelation prior to droplet formation. In the case of very fast-gelling hydrogel precursors, resulting microgels may retain a heterogeneous, two-halved arrangement, as shown in the inset. The scale bars denote 100 µm.

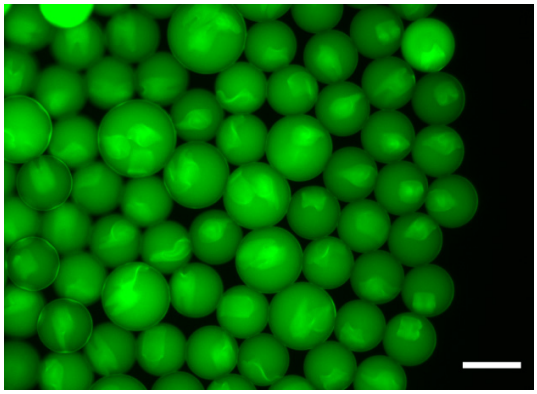

Figure S2: Coalesced water-in-oil droplets containing more than one anisotropic microgel. Droplets have coalesced due to the decreased solubility of the PEG block, that is part of the droplet-stabilizing PFPE-PEG-PFPE triblock copolymer, at high salt concentrations. This causes a reduced effectiveness of the surfactant. The scale bar denotes 100 µm.

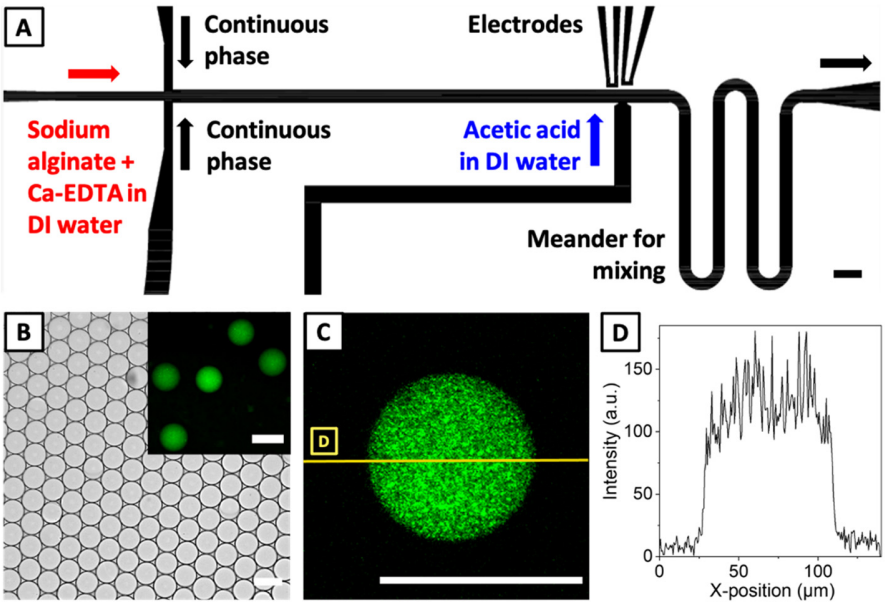

Figure S3: (A) AutoCAD®-made design of emulsion droplet electrocoalescence device, highlighting the inflow of the two hydrogel precursors, sodium alginate and calcium chloride. For fabricating isotropic alginate-based microgels via ionic interactions, DP1 was a uniform mixture of 2% (w/w) sodium alginate in an aqueous Ca-EDTA complex solution (0.05 M), and DP2 a 0.25% (v/v) solution of acetic acid in DI water. The Ca-EDTA complex solution was obtained by mixing a solution of calcium chloride (0.1 M) with a solution of disodium-EDTA (0.1 M) at equal volumetric ratios to achieve a final concentration of 0.05 M. Subsequently, the pH of the solution was adjusted to 7 using sodium hydroxide (5 M). For visualizing the mixing efficiency inside droplets, 0.2% (w/w) FITC-dextran (M_W: 2,000,000 g mol^{-1}) was added to DP1. After forming droplets of DP1 at the flow-focusing junction, DP2 was added into droplets of DP1, applying an electrode voltage of 240 V at 7.5 kHz in AC mode. Flow rates were set to 40 µL h^{-1} for DP1, 70 µL h^{-1} for DP2, and 400 µL h^{-1} for the CP. (B) Bright-field microscopy image of resulting emulsion droplets containing microgels that are formed *via* ionic interaction. The inset displays the corresponding fluorescence microscopy image of FITC-dextran entrapped inside the hydrogel network. (C) Exemplary confocal microscopy image of a microgel revealing the homogeneous distribution of fluorescent FITC-dextran. (D) Localization of FITC-dextran inside of the microgel shown in (C) visualized through a mid-plane line scan. The scale bars for all panels denote 100 µm.