Simplified Mesofluidic Systems for the Formation of Droplets and the Synthesis of Materials

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Reagents. 1,6-hexanediol diacrylate (HDDA, Aldrich) and ethylene glycol dimethacrylate (EGDMA, Fluka) were filtered through basic alumina before use to remove the inhibitor. Polyethylene imine (PEI, branched, MW ~ 1800, Polysciences), 2,2-dimethoxy 2-phenyl acetophenone (DMPA, Acros), carboxy methyl cellulose (CMC; M_w = 90,000; degree of substitution ~0.7; Acros), polystyrene (PS; linear; M_w = 350,000; M_n = 170,000; Aldrich), poly[methylene (polyphenyl) isocyanate] (PMPPI, Polysciences), Tween 80 (Aldrich), 95% ethanol (Mallinckrodt), chloroform (Mallinckrodt) and ethyl acetate (Mallinckrodt) were used as received. For the biphasic microparticle experiments, two dyes, rhodamine-6G (Acros) and coumarin 503 (Lambda Physik), were added to the monomer mixtures at a concentration of approximately 1 mg mL^{-1}.

Microfluidic Device Components. The channels consisted of 1/16" (1.588 mm) inner diameter (ID) polyvinylchloride tubing (VWR International Select Grade) and fluid junctions were made via 18, 20, and 30 gauge (838 μm ID/1,270 μm OD, 584 μm ID/902 μm OD, 154 μm ID/305 μm OD) blunt-end needles (Integrated Dispensing Systems). Holes were pierced into the channel walls prior to the insertion of the fluid needles by needles of the same size. Continuous phase fluids were contained in 20 mL syringes (Norm-Ject, latex-free) and the disperse phase was contained in 1 mL syringes (Norm-Ject, tuberculin). All fluids were driven with syringe pumps (Harvard Apparatus Model 22). Tubing connections were made with polypropylene fittings of several varieties, including male Luer-to-barb, female Luer-to-barb, barb-to-barb, and barbed Y junctions (Upchurch Scientific, all barbs for 1/16" tubing except for a barb-to-barb 3/8"-to-1/4" fitting) or stainless steel 20 gauge-to-16 gauge (584 μm ID/902 μm OD to 1,194 μm ID/1,651 μm OD) tubing adapters (Small Parts, Incorporated). A handheld ultra-violet (UV) lamp (Spectroline model ENF-260C) was used to cure the photopolymerizable monomers.

Janus-Faced Acrylate Droplets. A solution consisting of neat HDDA 4 mole % of DMPA as a photoinitiator was used as the disperse phase. The monomer solution was divided into two equal portions and the dyes, rhodamine 6G and coumarin 503, were added one dye to each portion (1 mg/mL). The two different colored monomer solutions were drawn into separate syringes (1 mL) with barbed Luer-lock fittings and both syringes were connected to a Y-junction. The Y-junction was fitted to a single needle that was connected to the FFD using the jig. This dual monomer solution became the disperse phase. The continuous phase of the FFD was an aqueous solution containing 1.5% CMC and 2.5% Tween 80 both by weight. By flowing the continuous phase at 0.5 mL/min (for each of the two syringes entering the FFD) and 10 μL/min for each of the two syringes containing the disperse phase, Janus-faced droplets were produced. This flow

rate is predicted to provide a Reynolds Number of 1 or less. The Janus-faced droplets were the photopolymerized using a handheld UV lamp using the 365nm setting (Spectoline model ENF-260C). The cured microparticles were then collected from the end of the tube and isolated by successive washing with DI water, 95% ethanol and finally ethyl acetate to remove the continuous phase and unreacted monomers.

Microparticle Concentrator. The 1/16" ID / 3/16" OD outlet tube from the FFD was fed through the *inside* of the 1/4" end of a polypropylene 1/4"-3/8" barbed adapter. A short (~ 4 cm) section of 3/8" ID PVC tubing was pushed onto the 3/8" barb end of the adapter and the other end of this tubing was connected to another 1/4"-3/8" barbed adapter (see Figure S1). The waste outlet channel was pushed into the inside of 1/4" end of the second adapter, similarly to the outlet from the FFD. To add the side channel, an 18 G needle was punctured into the bottom of the section of 3/8" ID PVC tubing. This large-gauge needle was connected to the side channel with a male Luer-to-barb fitting. To make the droplet-in-microcapsule, the outlet of the side channel was interfaced with a second continuous phase through a 20 G needle, attached to the side channel with another male Luer-to-barb fitting.

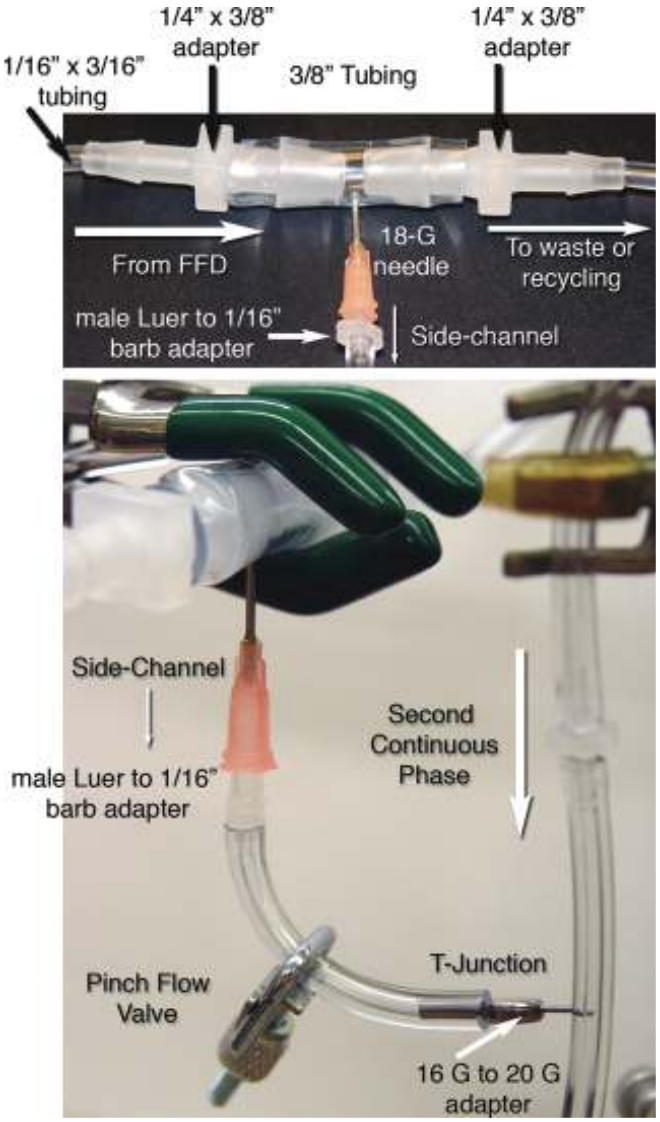

Figure S1. Photographs of the particle concentrator device and side channel with fittings and tubing labeled.

Characterization. The microparticles were examined with an optical microscope (Leica DM IL) and micrographs were captured with a digital camera (Sony DSC-F717) on a T-mount. Dark field fluorescence imaging was obtained with the same optical microscope by illuminating the microparticles with a broadband UV source (Leica mercury lamp) through the objective system and using filters appropriate to the dyes in the microparticles. Particle size distributions were determined by analyzing optical micrographs with Image J (available free from the National Institutes of Health at http://rsb.info.nih.gov/ij/) and statistical analyses were performed with Microsoft Excel. High speed imaging of the droplets was obtained with a high speed camera (Phantom 7.1, Vision Research, Inc.) through a camera lens (Nikkor F macro lens 105 mm, f/2.8, Nikon) illuminated from behind by a slide projector (Ektagraphie III AM, Kodak). Laser scanning confocal microscopy was performed on a Leica TCS SP2 microscope with a 10x dry objective.