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SUPPORTING INFORMATION

Continuous Synthesis of Elastomeric Macroporous Microbeads

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S1-Flow-Focusing Microreactor

Figure S1 shows the schematic of the off-the-shelf fittings, capillaries, and fluidic connections utilized for constructing the coaxial flow-focusing microreactor.



Figure S1. The capillary-based flow focusing microreactor constructed using off-the-shelf components. The bottom inset shows a bright-field image of the coaxial inner capillaries inserted inside the outer capillary.

Inlet-side T-junction (Left) connections listed clockwise from the left:

Polymer/crosslinker inlet line: 1 mm OD \times 0.75 mm ID \times 4 cm long inner glass capillary inserted into 1/16'' OD 0.04'' ID FEP tubing and threaded through the long axis of the T-junction.

Solvent/catalyst inlet line: 1/16'' OD FEP tubing connected to the perpendicular inlet of the T-junction.

Outlet of first T-junction: 1.5 mm OD \times 1.12 mm ID \times 6 cm long outer glass capillary with 1/8" OD x 1/16" ID Tygon sleeve fastened coaxially around inner capillary as it protrudes from the outlet of the T-junction.

Outlet side T-junction (Right) connections listed clockwise from the left:

Inlet of second T-junction: Other end of 1.5 mm OD \times 1.12 mm ID x 6 cm long outer glass capillary with another 1/8" OD \times 1/16" ID Tygon sleeve to fasten to the long axis of the outlet T-junction.

Continuous phase inlet line: 1/16" OD FEP tubing connected to the perpendicular inlet of the T-junction.

Reactor outlet: 1 mm OD \times 0.75 mm ID \times 10 cm long flamed-tip glass capillary with 1/16'' OD 0.04'' ID FEP sleeve fastened such that the two inner capillaries are separated by \sim 1.5 mm.

S2-Non-Porous Microbeads (Control)

Figure S2 shows the high degree of monodispersity obtained for the crosslinked nonporous PHMS microbeads (3:1 PHMS:hexadiene ratio) synthesized using the capillary-based microfluidic platform described in the paper.



Figure S2. Bright-field monolayer image of the PHMS microbeads synthesized using the developed 3D flow-focusing microreactor with PHMS:hexadiene ratio of 3:1.

S3-Silica-Loaded PHMS Microbeads

Figure S3 shows the high degree of monodispersity obtained for the (crosslinked) silicaloaded PHMS microbeads (5 μ m silica microspheres, 5:1 PHMS:hexadiene ratio) synthesized using the capillary-based microfluidic platform described in the paper.



Figure S3. Bright-field monolayer image of the silica-loaded PHMS microbeads synthesized using the developed 3D flow-focusing microreactor with 5µm silica microspheres and PHMS:hexadiene ratio of 5:1.

S4-Example Rheometer Compression Data

Figure S4 shows an example of data obtained for a non-porous PHMS microbead (control) from the compression test using the rheometer in parallel plate configuration.



Figure S4. Measured normal force *vs.* the parallel plate gap for an in-flow synthesized dense PHMS microbead with PHMS:hexadiene ratio of 3:1. The overlay of the compression/relaxation curves demonstrate the elasticity of the PHMS microbeads without hysteresis.

Supplementary Videos:

Video M-1 The droplet production videos of the flow-focusing microreactor for a porogen-free control and a silica-loaded test using a high-speed camera. Porogen-free flow parameters: $Q_1 = 10 \mu$ L/min, $Q_2 = 5 \mu$ L/min, and $Q_3 = 200 \mu$ L/min with 5:1 PHMS:hexadiene ratio. Silica-loaded flow parameters: $Q_1 = 10 \mu$ L/min, $Q_2 = 10 \mu$ L/min, and $Q_3 = 200 \mu$ L/min with 5:1 PHMS:hexadiene ratio and 10 μ m silica microspheres.

Video M-2 Reconstructed MicroCT volume renderings of multiple microbeads and single microbead sweep cut for the silica-loaded and macroporous PHMS microbeads. Microbeads were produced using $Q_1 = 10 \,\mu$ L/min, $Q_2 = 10 \,\mu$ L/min, and $Q_3 = 300 \,\mu$ L/min with 5:1 PHMS:hexadiene ratio and 5 μ m silica microspheres.