**Giant Volume Change of Active Gels under Continuous Flow**

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**Supporting Information**

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S1 Fabrication of microgels

Materials

2-hydroxy-4’-(2-hydroxyethoxy)-2-methylpropiophenone (Irgacure 2959), N,N’-methylene-bis-acrylamide (BIS, 5), N-isopropylacrylamide (NIPAAm, 4) (≥99%) were purchased from Sigma-Aldrich. NIPAAm was purified by recrystallization before use. All the ruthenium catalysts 1, 2 and 3 were synthesized as reported in previous publications.1

Fabrication of Gela microgels

NIPAAm (4, 40 mg), BIS (5, 0.7 mg) and Ru catalyst (1, 3.8 mg) were charged into a 0.5 mL eppendorf. Methanol (190 μL) and water (50 μL) were added into the eppendorf and the mixture was mixed well. Methanol solution of Irgacure 2959 (10 μL, 10 mg/mL) was added into the mixed solution. The mixture was injected into a polycarbonate-glass mold and sealed. After applying a mask on the glass side of the mold, the set-up was immersed in a shallow ice-water bath and exposed under an Omicure light source for 10 mins. The microgels were detached from the mold and immersed in DI water for dialysis for 3 days before further tests.

Fabrication of Gelb microgels

Fabrication of Gelb microgels is the same as the fabrication of Gela microgels, except that Ru catalyst (2, 3.6 mg) instead of Ru catalyst 1 was added to the reaction mixture.

Fabrication of Gelc microgels

NIPAAm (4, 40 mg) and Ru catalyst (3, 2 mg) were charged into a 0.5 mL eppendorf. Then the same procedure as in the fabrication of Gela and Gelb was followed. The exposure time to Omicure light source was extended to 30 min. After dialysis, Gelc microgels were obtained for further tests.
**Figure S1.** Optical image of the fabrication set-up with microgels inside (black circle part).

**S2 Characterization of the microgels**

**Figure S2.** (A) Optical image of a batch of microgels (Gel_a) (scale bar is 5 mm) before detachment from the glass slide and (B) an extended image (scale bar is 500 μm).

**Figure S3.** Fluorescent images of thin slice of microgels, (A) Gel_a, (B) Gel_b and (C) Gel_c (scale bar is 10 μm).

To determine the concentrations of the catalysts within the gels, we used a UV-vis spectrophotometer to measure the absorption of catalysts at $\lambda = 450$ nm, which helps to quantify the amount of unreacted catalysts monomer. By measuring the weight of the dried gel polymerized from a given volume of prepolymerization solution, we calculate the polymerization rate of NIPAAm (trace amount of BIS in Gel_a and Gel_b is ignored). Based on these data, we estimate that the polymerization conversion of NIPAAm in Gel_a is around 14% with a molar ratio of Ru-catalyst (1)/NIPAAm of 0.6%; the polymerization conversion of NIPAAm in Gel_b is around 23% with a molar ratio of Ru-catalyst (2)/NIPAAm of 0.5%, and the polymerization conversion of NIPAAm in Gel_c is around 11% with a molar ratio of Ru-catalyst (3)/NIPAAm of 5%.
**Figure S4.** Optical images of gels in the reduced state before and after several hours of BZ reaction. Gel_a at (A) 0 and (D) 6 h of BZ oscillation; Gel_b at (B) 0 and (E) 4 h of BZ oscillation; Gel_c at (C) 0 and (F) 6 h of BZ oscillation (scale bar is 100 μm).

**S3 Setup of microfluidic flow cells**

![Microfluidic flow cell setup](image)

**Figure S5.** Cartoon illustration of the set-up of the microfluidic flow cell for detection of chemomechanical oscillation of the microgels under a bright field microscope and on a temperature control stage.

**S4 Volume change of Gel_a under different conditions**

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Figure S6. The relative volume change of the oscillating Gel_a at same continuous-flow rate (10 μL/min) in the same microfluidic channel but different BZ reactant (listed in the table) at 10 °C.

S5 Volume change profiles of microgels under the same BZ flow at different temperatures

Figure S7. Oscillating volume change profile of Gel_a under a flow of BZ solution (10 μL/min, [NaBrO_3] = 0.2 M, [CH_2(COOH)_2] = 0.4 M, and [HNO_3] = 0.4 M) at (A) 10 °C, (B) 14 °C, (C) 18 °C, (D) 22 °C, (E) 26 °C and (F) 30 °C.

Figure S8. Oscillating volume change profile of Gel_b under a flow of BZ solution (10 μL/min, [NaBrO_3] = 0.2 M, [CH_2(COOH)_2] = 0.4 M, and [HNO_3] = 0.4 M) at (A) 14 °C, (B) 22 °C and (C) 30 °C.
Figure S9. Oscillating volume change profile of Gelc under the flow of BZ solution (10 μL/min, [NaBrO₃] = 0.2 M, [CH₂(COOH)₂] = 0.4 M, and [HNO₃] = 0.4 M) at 10 °C (A), 14 °C (B) and 18 °C (C).

S6 Volume change profiles of the microgels under continuous flow and stationary conditions

![Graphs](image1)

Figure S10. Oscillating volume change profiles of (A) Gelₐ, (C) Gelₕ and (E) Gelc with continuous flow, (B) Gelₐ, (D) Gelₕ and (F) Gelc without flow under optimal BZ conditions.

S7 Profiles of the volume change of Gelₐ at different flow rates of the BZ solutions in microfluidic channels having different widths
Figure S11. Time dependent relative diameter profiles of Gel a in a microfluidic channel with a width 40 times the diameter of Gel 1 at a flow speed of (A) 10 µL/min, (B) 50 µL/min, and a channel with a width 10 times the diameter of Gel a at a flow speed of (C) 2.5 µL/min, (D) 5 µL/min and (E) 10 µL/min. (w, width of microfluidic channel; d, diameter of microgel.)

S8 Volume change profiles of Gel a during counter ion exchange

Figure S12. Relative volume change profiles of Gel a at same continuous-flow rate (10 µL/min) but with counter ion exchange (A) from HNO₃ to H₂SO₄ and (B) from H₂SO₄ to HNO₃.

Movie S1 Volume change of Gel b under continuous flow of BZ solution in the microfluidic channel
Movie S2 Volume change of array of Gelₐ under continuous flow of BZ solution in the microfluidic channel