Electronic Supplementary Material (ESI) for ChemComm.

Electronic Supplementary Information

Formation of a core-shell droplet in a thermo-responsive ionic liquid/water mixture by optical tweezers

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S1. Sample preparation

The ionic liquid was prepared from tetrabutylphosphonium hydroxide (40 w/w% in water) (TCI) and *m*-xylene-4-sulfonic acid (solid, purity: >98.0%) (TCI). Equimolar amounts of them were mixed in a sample tube. The mixture was heated on a hotplate, evaporating the water. The remaining IL was diluted with an appropriate amount of water to adjust the concentration.

S2. Optical setup

A continuous-wave (cw) 1064 nm laser beam (Shanghai Laser & Optics Century, IR 1064T5-3000) was used for the optical tweezers. The laser was introduced into an inverted microscope (Nikon, ECLIPSE Ti2) and focused onto a sample solution sandwiched between glass slides through an objective lens (Nikon, ×100, oil, N.A.=1.45). The diameter of the trapping laser was estimated to be 0.90 μm at the focal spot. The optical images were captured with an imaging camera (Omron Sentec, STC-MVS322U3V). For confocal Raman microspectroscopy, a cw 532 nm laser (HC

Photonics) was used as the excitation light source. The scattered light was guided to a pinhole with a diameter of 150 µm. Then, the light was introduced to a spectrograph (SOL instruments) and detected with a charge coupled device (CCD) detector (Andor). Fluorescence images and spectra were obtained under the irradiation of a cw 405 nm laser (Backer & Hickl GmbH, BDS-SM-405-FBE).

S3. Release of the droplet in the prolonged irradiation

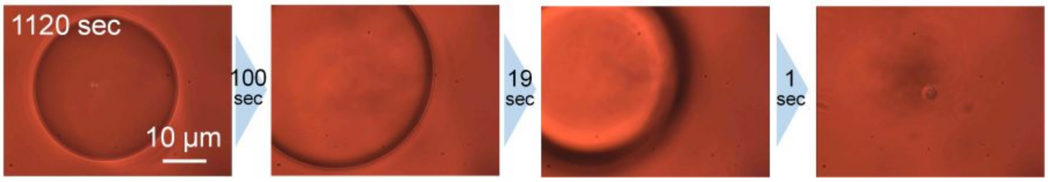

Fig. S1 Optical transmission micrographs around the focal spot under the prolonged laser irradiation at 630 mW. The sample is different from that of Fig. 2a.

S4. Laser power dependence on the final droplet size

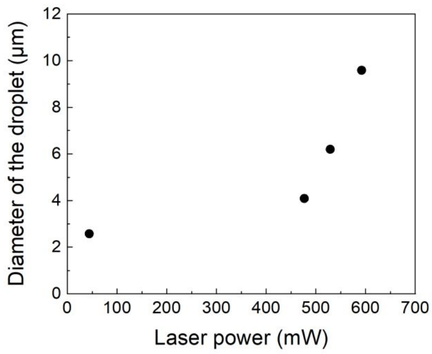

Fig. S2 The relation of the size of the final microdroplet and the input laser power.

S5. Stability of the core

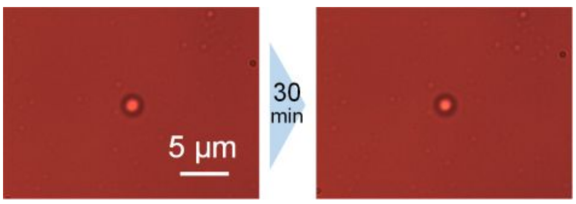

Fig. S3 Optical transmission micrographs of the core on a bottom glass substrate. The core was moved to the substrate by lowering the focal position and observed for 30 min without the laser irradiation.