Supporting Information for Lab on a Chip

Controllable Microfluidic production of Gas-in-Oil-in-Water emulsions for hollow microspheres with thin polymer shells

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Microfluidic device: Cross-junction channels are fabricated on a polymetyl methacrylate (PMMA) plate using a Computerized Numerical Control (CNC) machine tool with an end mill (Φ=1mm). The plate is then similarly cut into several chips (35mm×20mm×3mm) each with a cross-junction channel. The channel for dispersed phase fluid is approximately 1.5 mm wide × 1.5 mm high, the channel for continuous phase is approximately 1.35 mm wide × 1.35 mm high. A circular glass capillary with inner-diameter of 0.3 mm and outer-diameter of 0.6 mm is tapered using a micropipet puller (P-97, SUTTER Co. Ltd., USA) for the injection of the gas phase fluid. The diameter of the tapered orifice is approximately 9 μm. The first tapered capillary is inserted into another capillary with inner-diameter of 0.86 mm and outer-diameter of 1.5 mm for the middle phase and the orifice is also tapered to approximately 60μm. We make sure that both the orifices align in the same plate. Then, those compound capillaries are inserted into a third coaxially aligned capillary for the double emulsions. The tolerance required for alignment of the capillaries is 5μm. The inner-diameter of the third capillary is 0.86 mm and the outer-diameter is 1.5 mm. PTFE pipes are inserted into the channel and capillaries and microsyringe pumps (LSP01-1B, Baoding Longer Precision Pump Co., Ltd) are used to pump the gas phase and two liquid phases into the microfluidic device respectively.

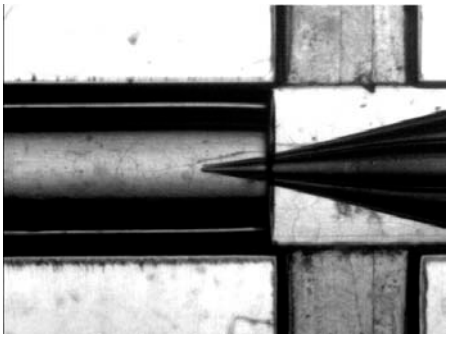

Fig. S1 The micrograph of capillary microfluidic device.

Materials and methods: For the flow-rate dependent measurements, we dispersed air bubbles into silicone oil with the viscosity of 10mPa·s (Jing Pinghua Co. Ltd). The oil droplets were then dispersed into deionized water with 2 wt. % PVA (average molecular weight: 1788, Aladdin chemistry Co.Ltd). For the polymerization reactions, 99% of 1,6-Hexanediol diacrylate (Aldrich) and 1% of photoinitiator 2-Hydroxy-2-methylpropiophenone (Aldrich) was used as the oil phase. The HDDA droplets encapsulating bubbles were polymerized by activating the photo-initiator using a UV spot light source (Hamamatsu, Model L9588-01). All the reagents are of analytical grade and used as received. The formation of the droplets and bubbles was carried out with an optical microscope (BX61, Olympus, Japan) equipped with a high-speed camera with a frequency

of 200 images per second (B742, Pixelink, Canada). More detailed structures were observed using scanning electron microscopy (SEM, FEI XL30). Fluorescence was observed using laser scanning confocal microscopy (LSM710, Zeiss). The UV spot light source (Hamamatsu UV Spot Light Source L9588-01 Lightning cure) is operated under the intensity of 100% and wavelength of 365nm. All the experiments were carried out at room temperature (25^{o}C).

Characterization

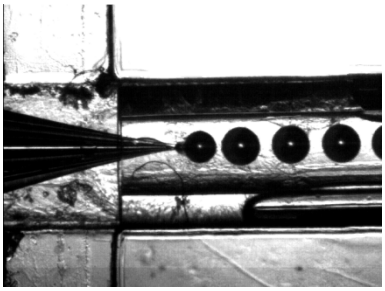

Fig. S2 The typical micrographs of G/O/W emulsion droplet formation in the microfluidic device.

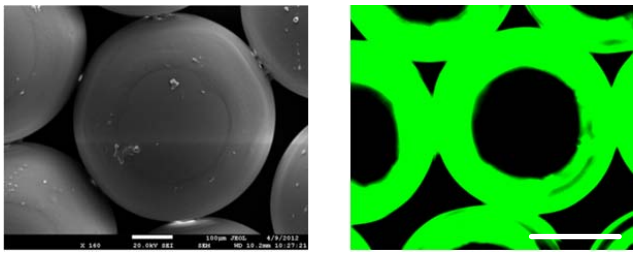

Fig. S3 The SEM and LSCM photograph of the hollow microspheres with relatively thick shell. The scale bar is 350μm.