Supplementary Information

Microfluidic fabrication of complex shaped microfibers by liquid template-aided multiphase microflow

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1. Configuration and notations of the englufing for the generation of hollow microfibers

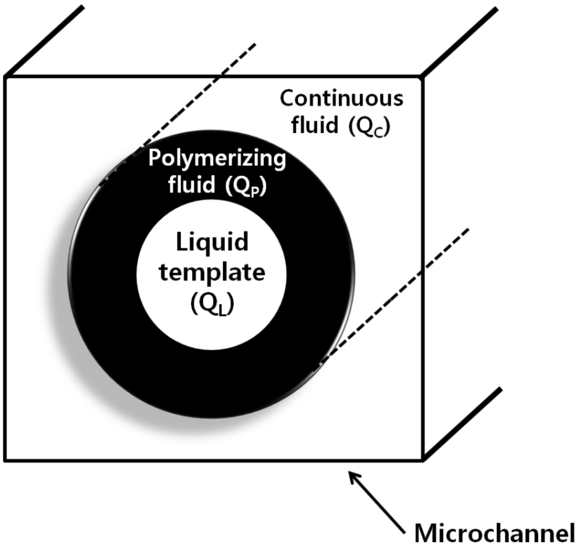

Fig. S1 Schematic diagram describing cross-sectional view of core-shell jet and illustrating immiscible continuous fluid (outer phase), polymerizing fluid (middle phase), and liquid template (inner phase) in the microchannel.

2. Compositions and physical characteristics of fluids at the case of formation of engulfing flow

Table. S1 Compositions and physical characteristics of each phase used for formtion of engulfing flow

Phase	Composition	Density (g/ml)	Viscosity at 20℃	Interfacial tension (mN/m)	Spreading coefficient*
Liquid template (Q_L)	100% PEGDA (M_n=575)	1.1	55	γ_{LC} = 2.9 ± 0.02	S_L =-2.3
Polymerizing fluid (Q_P)	50 wt% PEGDA,42wt% water and 8 wt% Darocur1173	0.78	15	γ_{LP} = negligible	S_P =2.9
Continuous fluid (Q_C)	95 wt% Hexadecane and5 wt% span 80	1.1	8	γ_{PC} = 0.6 ± 0.01	S_C =-3.5

*Spreading coefficient are calculated by $S_L = \gamma_{PC}-(\gamma_{LP}+\gamma_{LC})$; $S_P = \gamma_{LC}-(\gamma_{LP}+\gamma_{PC})$; $S_C = \gamma_{LP}-(\gamma_{LC}+\gamma_{PC})$.

3. Thermodynamic values among three phases for the prediction of microfiber geometries

There are three possible configurations in the equilibrium state: when S_L is negative value, (i) conditions induce core–shell (completely engulfing) morphology, with the liquid template fluid appearing as the core within a shell of polymerizing fluid; (ii) conditions correspond to stratified jetting; and (iii) conditions describe separated jetting.

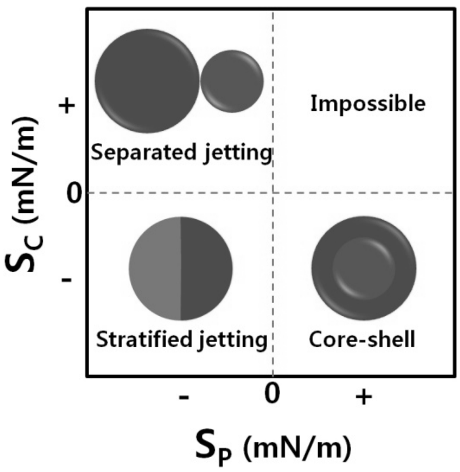

Fig. S2 Schematic diagram of possible configurations corresponding to each spreading coefficient (S_P and S_C) when S_L is always negative value.

Table. S2 Interfacial tensions measured at various experimental conditions of fluids. The estimated spreading coefficient providies information of prediction of microfiber morphologies

Q_C (continuous fluid)	Q_P (polymerizing fluid)	Q_L (liquid template)	Interfacial tension (mN/m)	Spreading coefficient	Estimated Morphology	Observed Morphology
95 wt% hexadecane with 5 wt% span 80	50% PEG-DA, 42wt% water and 8wt% DEAP	100 wt% PEG-DA	γ_{LP} = negligible γ_{LC} = 2.9 ± 0.02 γ_{PC} = 0.6 ± 0.01	$S_L < 0, S_P > 0, S_C <$	Engulfing flow (hollow microfiber)	Engulfing flow (hollow microfiber)
		80 wt% PEG-DA	γ_{LP} = negligible γ_{LC} = out of range γ_{PC} = 0.6 ± 0.01	$S_L \leq 0, S_P \geq 0, S_C <$	Engulfing flow (hollow microfiber)	Engulfing flow (hollow microfiber)
		60 wt%	γ_{LP} =	$S_L < 0, S_P > 0, S_C <$	Engulfing flow	Engulfing flow

		PEG-DA	negligible γ_{LC} = 0.9 ± 0.45 γ_{PC} = 0.6 ± 0.01 γ_{LP} = negligible		(hollow microfiber)	(hollow microfiber)
		40 wt% PEG-DA	γ_{LC} = 1.8 ± 0.03 γ_{PC} = 0.6 ± 0.01 γ_{LP} = negligible	$S_L < 0, S_P > 0, S_C <$	Engulfing flow (hollow microfiber)	Engulfing flow (hollow microfiber)
		20 wt% PEG-DA	γ_{LC} = 2.6 ± 0.07 γ_{PC} = 0.6 ± 0.01 γ_{LP} =negligibl e	$S_L < 0, S_P > 0, S_C <$	Engulfing flow (hollow microfiber)	Engulfing flow (hollow microfiber)
		0 wt% PEG-DA (water)	γ_{LC} =5 $.2 \pm 0.12$ γ_{PC} = 0.6 ± 0.01 γ_{LP} = negligible	$S_L < 0, S_P > 0, S_C <$	Engulfing flow (hollow microfiber)	Engulfing flow (hollow microfiber)
	92% PEG-DA, 8wt% DEAP	100 wt% PEG-DA	γ_{LC} = 2.9 ± 0.02 γ_{PC} = 2.5 ± 0.12	$S_L \lesssim 0, S_P \lesssim 0, S_C$	Stratified Jetting	Stratified Jetting

4. The evolution of hollow microfiber geometries

We further provide more detailed results of hollow microfiber for the results shown in Figure 2 as shown below in Figure S2. The detailed information of evolution of hollow microfiber is investigated with sequential optical images and SEM imgaes. The increase of evolution time along the microchannel will place the cocentric core structure from excentered core region. In early evolution state (1st state), mechanical strength of the hollow fiber is relatively low because one side of fiber is much thinner than opposite side. From second state to third state, while polymerized hollow fiber retain own structures, the position of hole still is not placed in the center. Finally, symmetric hollow microfiber can be produced using fully developed engulfing jets at fourth state.

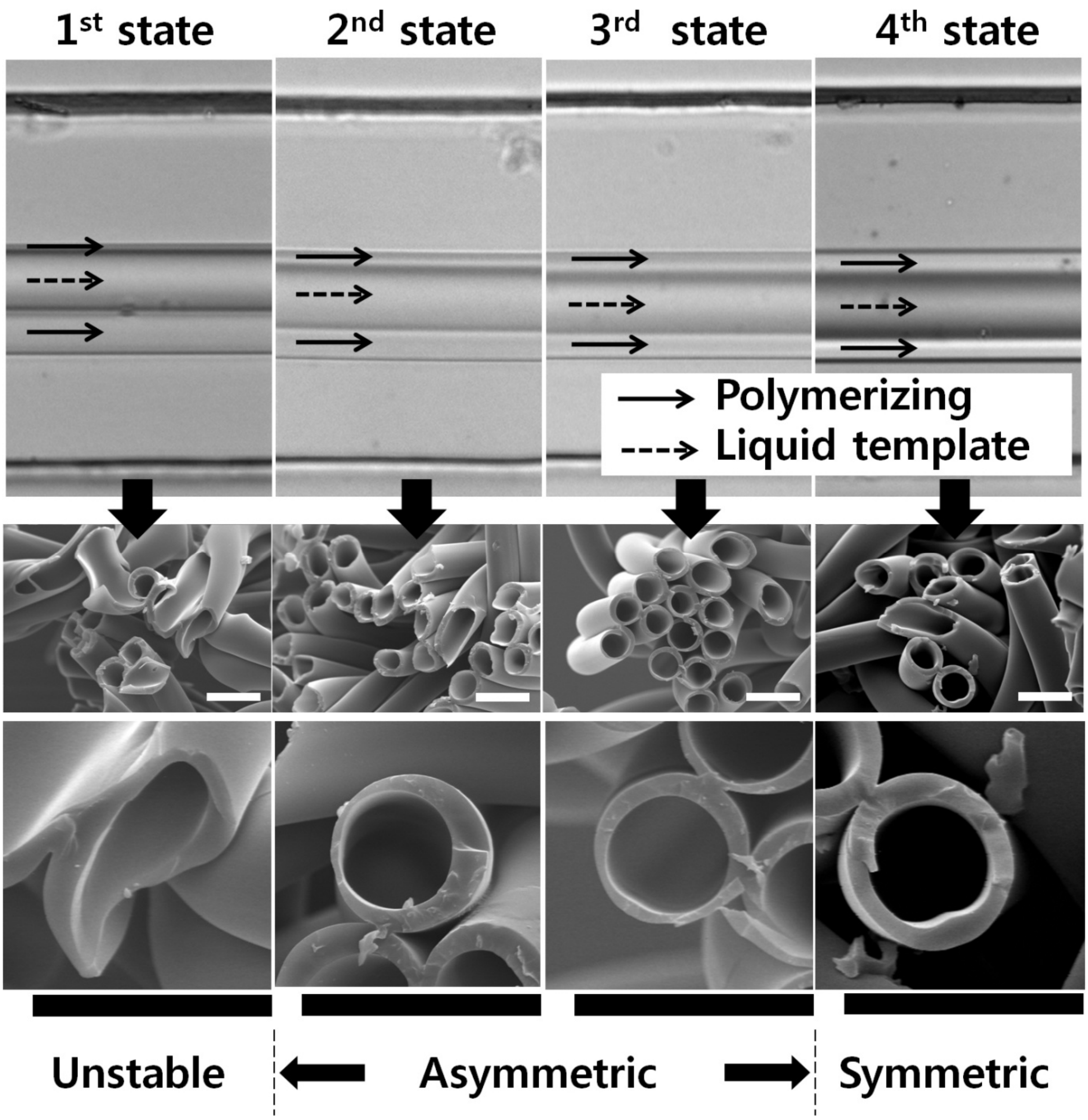

Fig. S3 Polymerized hollow microfibers at different evolution states upon UV exposure as shown in Figure 1B. Scale bars represent 50 μm.