Fabrication of Solvent Transfer-Induced Phase Separation Bijels with Mixtures of Hydrophilic and Hydrophobic Nanoparticles

AUTHORS NAMES: Giuseppe Di Vitantonio,[a] Daeyeon Lee*[a] and Kathleen J. Stebe*[a]

[a]Department of Chemical and Biomolecular Engineering, University of Pennsylvania,
Philadelphia, PA 19104, USA;

*To whom correspondence should be addressed
Figure S1: Schematics of STRIPS bijel manufacturing and collection.

Figure S2: SEM images of STRIPS bijels.

Movie S1: Confocal microscopy of STRIPS fiber with hydrophilic fraction below the optimal range.

Movie S2: Confocal microscopy of STRIPS fiber with hydrophilic fraction within the optimal range.

Movie S3: Confocal microscopy of STRIPS fiber with hydrophilic fraction above the optimal range.

Movie S4: Confocal microscopy of STRIPS fibers with different total nanoparticles concentrations but the same hydrophilic fraction.

Movie S5: Confocal microscopy of STRIPS fiber made with a DVB-BrPh oil phase with a hydrophilic fraction within the optimal range.

Movie S6: Confocal microscopy of STRIPS fiber made with a HDA-HxOH oil phase with a hydrophilic fraction within the optimal range.

Movie S7: Confocal microscopy of STRIPS fiber made with a HDA-HxO oil phase with a hydrophilic fraction within the optimal range.

Movie S8: Confocal microscopy of STRIPS fiber made with fluorescent nanoparticles.
Figure S1. Schematics for STRIPS bijel manufacturing and collection. The two syringes are connected to their corresponding nozzle using polyethylene tubing. Syringe pumps are used to pump the two solutions in the microfluidic device.
Figure S2. SEM images of STRIPS bijels. Cross sectional image of polymerized STRIPS bijels.
**Movie S1.** Confocal microscopy of STRIPS fiber with hydrophilic fraction below the optimal range.

The following materials are used: (i) 10 wt% CAB-O-SIL TS-610 in hexanediol diacrylate (HDA), (ii) 15 wt% CAB-O-SIL LM-150 in ethanol, (iii) 15 wt% CAB-O-SIL TS-610 in ethanol, (iv) pH 2.23 water solution, and (v) 2-hydroxy-2-methylpropiophenone (HMP). Components (i)-(v) are mixed with the following proportions: (i) 1.14 g, (ii) 0.41 g, (iii) 0.63 g, (iv) 0.428 g, (v) 0.030 ml. The resulting suspension is used for the STRIPS process after the addition of Nile red for fluorescence purposes. UV light is used to trigger the polymerization of the oil phase. Diethyl phthalate (DEP) is used for refractive index matching. DEP is used unless otherwise specified.

**Movie S2.** Confocal microscopy of STRIPS fiber with hydrophilic fraction within the optimal range.

The following materials are used: (i) 10 wt% CAB-O-SIL TS-610 HDA, (ii) HDA, (iii) 15 wt% CAB-O-SIL LM-150 in ethanol, (iv) 15 wt% CAB-O-SIL LM-150 in a pH 2.23 water solution, and (v) 2-hydroxy-2-methylpropiophenone (HMP). Components (i)-(v) are mixed with the following proportions: (i) 0.58 g, (ii) 0.63 g, (iii) 0.984 g, (iv) 0.5 g, (v) 0.030 ml. The resulting suspension is used for the STRIPS process after the addition of Nile red. UV light is used to trigger the polymerization of the oil phase.
**Movie S3.** Confocal microscopy of STRIPS fiber with hydrophilic fraction above the optimal range.

The following materials are used: (i) HDA, (ii) 15 wt% CAB-O-SIL LM-150 in ethanol, (iii) 15 wt% CAB-O-SIL LM-150 in a pH 2.23 water solution, and (iv) 2-hydroxy-2-methylpropiofenone (HMP). Components (i)-(iv) are mixed with the following proportions: (i) 1.02 g, (ii) 1.02 g, (iii) 0.5 g, (iv) 0.030 ml. The resulting suspension is used for the STRIPS process after the addition of Nile red. UV light is used to trigger the polymerization of the oil phase. The hydrophilic fraction is 1.

**Movie S4.** Confocal microscopy of STRIPS fibers with different total nanoparticles concentrations but the same hydrophilic fraction.

The following materials are used: (i) 10 wt% CAB-O-SIL TS-610 HDA, (ii) HDA, (iii) absolute ethanol, (iv) 20 wt% CAB-O-SIL LM-150 in ethanol, (v) 20 wt% CAB-O-SIL LM-150 in a pH 2.23 water solution, (vi) pH 2.23 water solution, and (vii) 2-hydroxy-2-methylpropiofenone (HMP). LM 150 nanoparticles fraction is kept constant at 80%, while the total nanoparticles fraction is changed as shown in Figure 4 in the main text. The top row fibers from left to right have 14.5% and 12% wt of silica nanoparticles respectively. The bottom row fibers from left to right have 10.4% and 8% wt of silica nanoparticles respectively.

**Movie S5.** Confocal microscopy of STRIPS fiber made with a DVB-BrPh oil phase with a hydrophilic fraction within the optimal range.

The following materials are used: (i) bromobenzene (BrPh), (ii) divinyl benzene (DVB), (iii) 10 wt% CAB-O-SIL LM-150 in ethanol, (iv) 14 wt% CAB-O-SIL TS-610 in ethanol, (v) pH 1.63 water solution, (vi) 2-hydroxy-2-methylpropiofenone (HMP). Components (i)-(vi) are
mixed with the following proportions: (i) 0.713 g, (ii) 0.713 g, (iii) 0.537 g, (iv) 0.563 g, (v) 0.11 ml, (vi) 0.07 ml. The resulting suspension is used for the STRIPS process after the addition of 9,10-Bis(phenylethynyl)anthracene. UV light is used to trigger the polymerization of the oil phase. This fiber is shown in Figure 3 in the main text. A 38 wt% DEP and 62 wt% mixture of cinnamaldehyde are used for refractive index matching.

**Movie S6.** Confocal microscopy of STRIPS fiber made with a HDA-HxOH oil phase with a hydrophilic fraction within the optimal range.

The following materials are used: (i) HDA, (ii) cyclohexanol (HxOH), (iii) 20 wt% CAB-O-SIL LM-150 in ethanol, (iv) 20 wt% CAB-O-SIL LM-150 in pH 2.5 water solution, (v) pH 2.5 HCl in water solution, (vi) 2-hydroxy-2-methylpropiophenone (HMP). Components (i)-(vi) are mixed with the following proportions: (i) 0.382 g, (ii) 0.364 g, (iii) 0.954 g, (iv) 0.582 g, (v) 0.311, (vi) 0.03 ml. The resulting suspension is used for the STRIPS process after the addition of Nile red. UV light is used to trigger the polymerization of the oil phase. This fiber is shown in Figure 3 in the main text.

**Movie S7.** Confocal microscopy of STRIPS fiber made with a HDA-HxO oil phase with a hydrophilic fraction within the optimal range.

The following materials are used: (i) HDA, (ii) cyclohexanone (HxO), (iii) 20 wt% CAB-O-SIL LM-150 in ethanol, (iv) 20 wt% CAB-O-SIL LM-150 in pH 2.5 water solution, (v) 2-hydroxy-2-methylpropiophenone (HMP). Components (i)-(v) are mixed with the following proportions: (i) 0.44 g, (ii) 0.44 g, (iii) 0.888 g, (iv) 0.824 g, (v) 0.03 ml. The resulting suspension is used for the STRIPS process after the addition of Nile red. UV light is used to trigger the polymerization of the oil phase. This fiber is shown in Figure 3 in the main text.
Movie S8. Confocal microscopy of STRIPS bijel fiber with fluorescent nanoparticles.

The following materials are used: (i) HDA, (ii) DEP, (iii) 10 wt% FITC-CAB-O-SIL LM-150 in ethanol, (iv) 10 wt% RITC-CAB-O-SIL TS-610 in ethanol, (v) pH 2.23 water solution, (vi) 2-hydroxy-2-methylpropiophenone (HMP). Components (i)-(vi) are mixed with the following proportions: (i) 0.5 g, (ii) 0.5 g, (iii) 0.684 g, (iv) 0.292 g, (v) 0.428 g, (vi) 0.03 ml. The movie on the left shows a bijel fiber made with fluorescent nanoparticles. FITC labeled nanoparticles are in green and RITC labeled nanoparticles are in red. Nile red is added in DEP and confocal scan is performed after the dye permeation too (right movie).