Electronic Supplementary Information (ESI)

High-Throughput Optofluidic Platforms for Mosaicked Microfibers toward Multiplex Analysis of Biomolecules

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S1. Experimental

Preparation of Fluorescent Dye Modified Silica Particles: Sol-gel based Stöber method was used to prepare monodisperse silica particles with the size of 330 nm. Fluorescent dye solutions were prepared by mixing fluorescent dye molecules with aminopropyl-trimethoxysilane (APMS, Aldrich); tetramethyl rhodamine isothiocyanate (TRITC, Sigma-Aldrich), fluorescein isothiocyanate (FITC, TCI) and coumarin 343 (Sigma-Aldrich)) were used. Then three dye solutions were added into 1% (w/w) ethanolic suspension of silica particles with tetraethyl orthosilicate (TEOS, Aldrich) and ammonium hydroxide (Sigma-Aldrich), respectively. This sol-gel reaction proceeded for 48 h. Further reaction with additional TEOS for 12 h led to formation of silica thin shells on bare dye tagged silica particles.

Acrylation of Immunoglubulin G: Human IgG (reagent grade,≥95%, buffered aqueous solution , Sigma-Aldrich) and rabbit IgG (technical grade,≥80%, buffered aqueous solution , Sigma-Aldrich) were acrylated by reacting with 5000 Da acrylate-PEG-NHS (Jenkem technology) at 1:1 molar ratio in 50 mM sodium bicarbonate buffer of pH 8. The reaction was run for 3 h at room temperature under continuous stirring. Dialysis were proceeded to remove unreacted acryl-PEG-NHS against deionized water using 5000 MWCO cellulose dialysis membrane (Spectra/por)

Preparation of solutions for mosaicked microfibers with fluorescence: A solution for core fluid was prepared in proportion of 95% (v/v) of poly(ethylene glycol)(575) diacrylate (PEGDA, Sigma-

Aldrich), 5% (w/w) of Darocur 1173 (Ciba Specialty Chemicals) and 0.5% (v/v) of fluorescent dye modified silica particles. Pure poly(ethylene glycol)(400) (PEG, USB Corporation) was used as carrier fluids.

Preparation of solutions for biomolecular analysis: The core fluid was formulated with 50% (v/v) of PEGDA, 5% (v/v) of Darocur 1173 and 0.1% (w/v) of IgG in 0.01 M phosphate buffered saline, (pH 7.4) (PBS, Sigma Aldrich). The solution of carrier fluid was prepared with 50% (v/v) of PEG in PBS.

Fabrication of microfluidic devices: Firstly, photomasks for microchannels were designed with AutoCAD. Then, conventional photolithography was conducted by using SU-8 photoresist (Microchem) to make channel patterns for both top and bottom layers. Subsequently, 10:1 mixture of polydimethylsiloxane (PDMS, Sylgard 184, Dow Corning) and base curing agent was poured over SU-8 microchannel patterns and cured in convection oven at 70℃ for 2 h. Finally, PDMS molds for top and bottom layers were detached from substrates and then, carefully combined after the oxygen plasma treatment.

Generation of microfibers in microfluidic devices: The microfluidic device was mounted on an inverted optical microscope (Eclipse TE 2000-U, Nikon). Then, each fluid was introduced to microchannels with various volumetric flow rates using syringe pumps (KDS 200, KD Scientific). After stabilizing the flow for a several minute, UV light was irradiated through a lens using super high pressure mercury lamp (100 W, C-SHG1, Nikon). A core flow was photopolymerized with the desired microfiber composition. Finally, a polymeric microfiber was collected at the outlet of microfluidic device.

Detection of target biomolecules using mosaic-patterned microfibers: Three sample solutions were prepared. Each solution was formulated with 20 μg/ml of anti-immunoglobulin G (anti-IgG) in

PBS buffer. Two types of anti-IgG, FITC-anti-human IgG (Sigma-Aldrich) and TRITC-anti-rabbit IgG (Sigma-Aldrich) were used. Microfibers were incubated with these three solutions for 30 min at 35℃ and washed with DI water for several times. A confocal laser scanning microscope (LSM 5 PASCAL, Zeiss) was used to detect fluorescent signals from target molecules and it processed images for Top, cross-sectional and bottom views.

S2. Manipulation of microfiber dimensions.

| Fractional volume of core liquid (ϕ_{core})	Flow rate (μl/hr)		
	Total	Core	Carrier
0.4	200	80	60
0.5	200	100	50
0.6	200	120	40
0.7	200	140	30
0.8	200	160	20

Table S1. Flow rates of core and carrier liquid flows corresponding to the fractional volumes of core liquid

flows (ϕ_{core}).

S3. Composition change of microfibers.

| $\phi_{red,flow}$	Flow rate (μl/hr)			
	Total	Carrier	Red core	Green core
0.1	500	100	40	360
0.3	500	100	120	280
0.5	500	100	200	200
0.7	500	100	280	120
0.9	500	100	360	40

Table S2. Fractional volumes of red fluorescent flow, $\phi_{red,flow}$, and flow rates of red and green cores, and carrier liquid flows,.

S4. Formation of stratified laminar flows
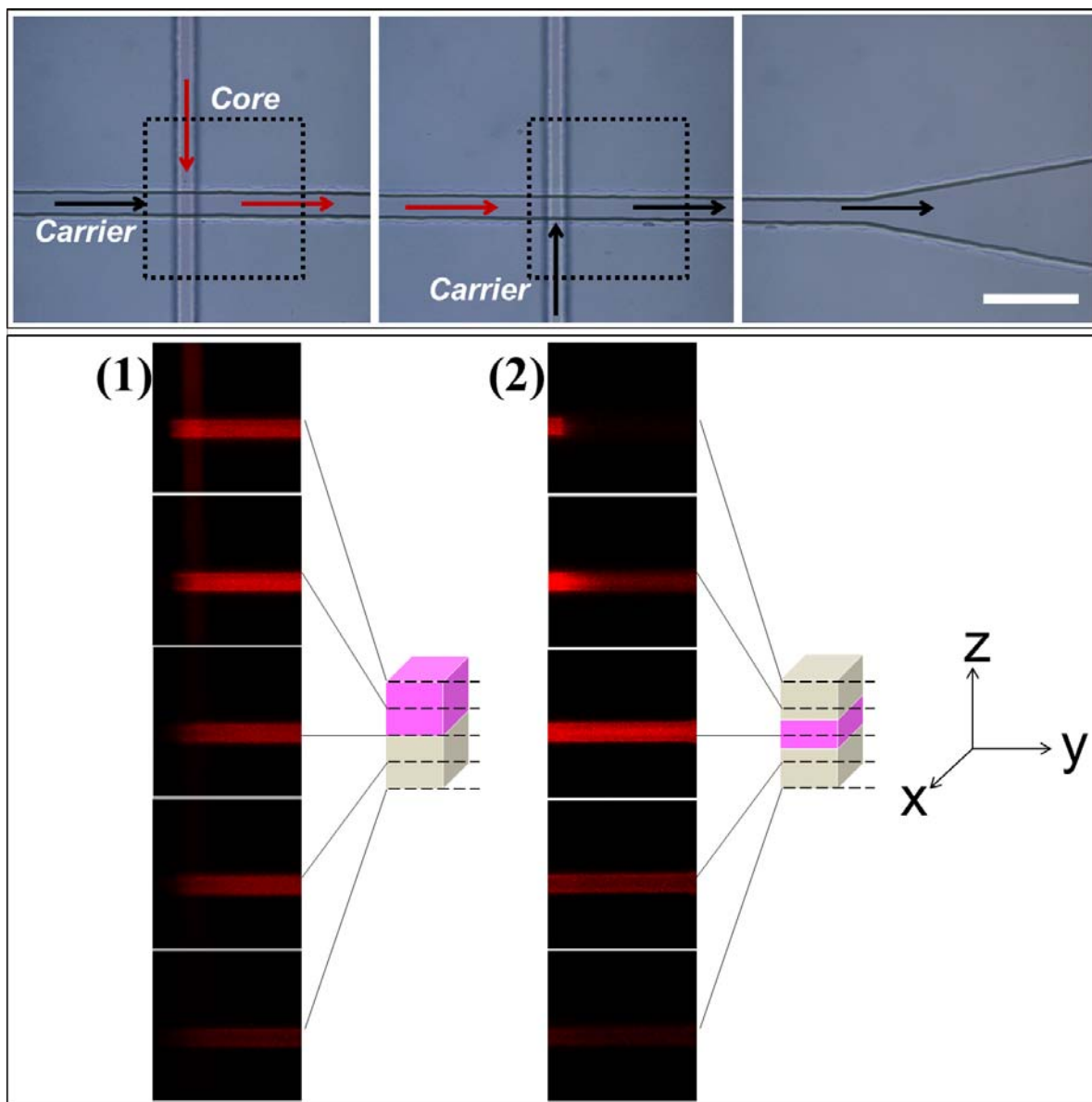
Fig. S1 Gallery of confocal microscope images obtained from cross junctions. Each cross junction was scanned in z-direction and visualized with red fluorescent dyes in a core flow. Formation of bilayer and trilayer laminar flows is represented. Scale bar for (A) is 200 μm.

S5. High-throughput generation of microfibers
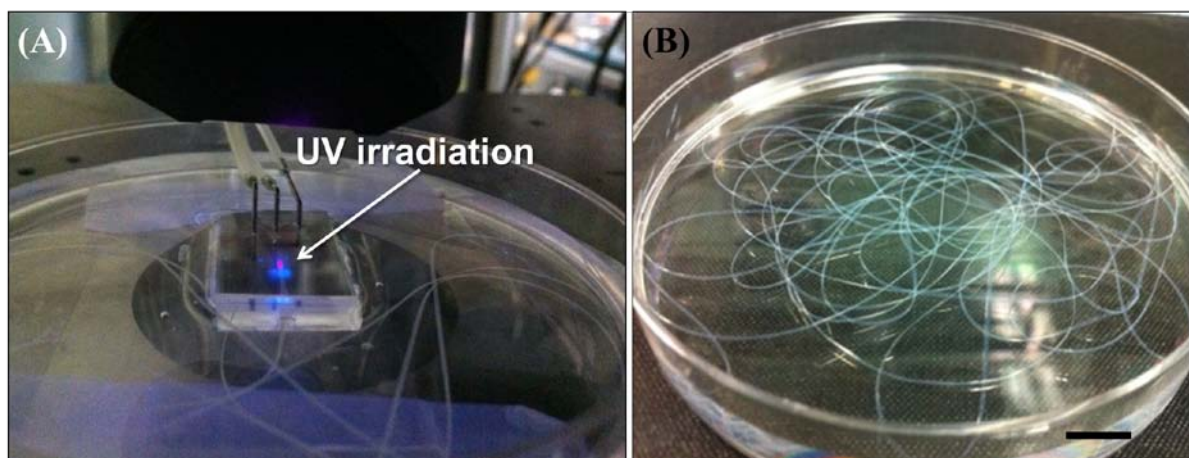
Fig. S2   Continuous generation of a microfiber in a microfluidic device (A) Digital camera image of the microfluidic device over UV irradiation. (B) A meter-long microfiber immersed in water. Approximately 3 m of microfiber was generated for more than a hour-long UV exposure. Scale bar is 1 cm for (B).
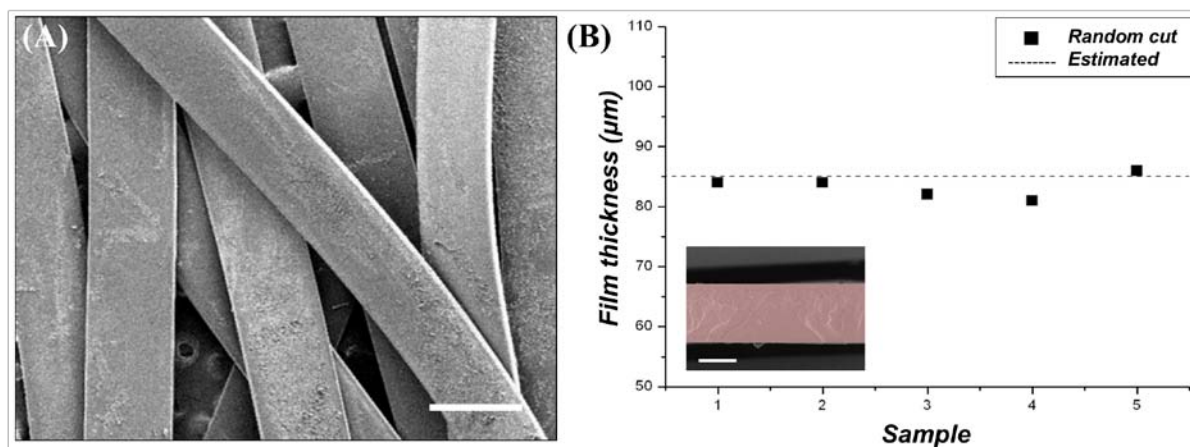
S6. SEM images of microfiber with different dimensions
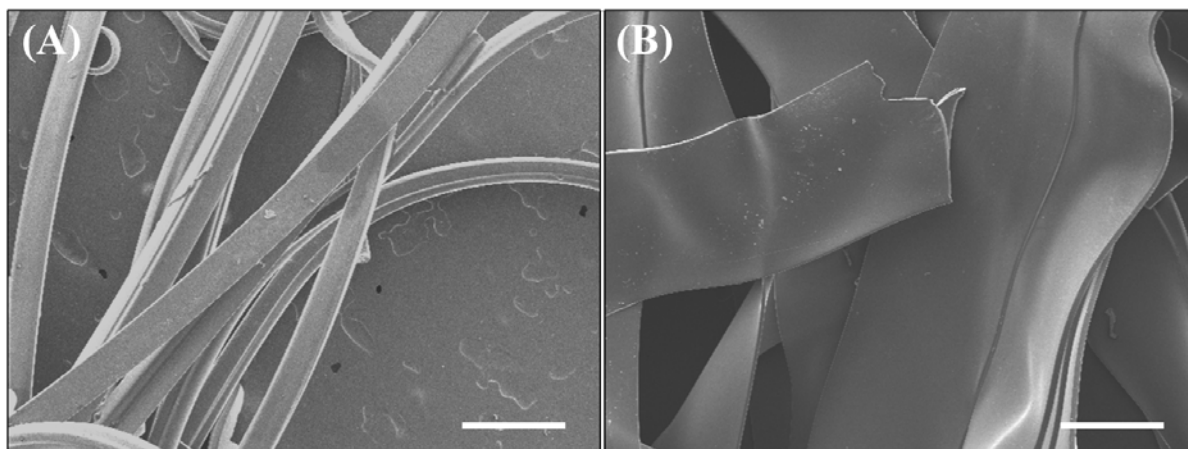
Fig. S3 Width tuning of microfibers by using different microchannels. (A) The SEM image of microfibers fabricated in microchannel of 200 μm and (B) microfibers fabricated in microchannel of 1000 μm. All scale bars are 500 μm.

S7. Uniform thickness of microfibers

Fig. S4   The uniformity of a microfiber (A) The SEM image of a microfiber with the uniform width. (B) Radom cuts of five pieces were collected and thickness was measured. The calculated microfiber thickness was 85 µm and the average of measured thickness was 83.4 µm with standard deviation of 1.95, which confirmed the uniformity of microfiber thickness. Scale bars are 500 µm for (A) and 50 µm for (B).

S8. Sensitivity of microfiber for biomolecular analysis
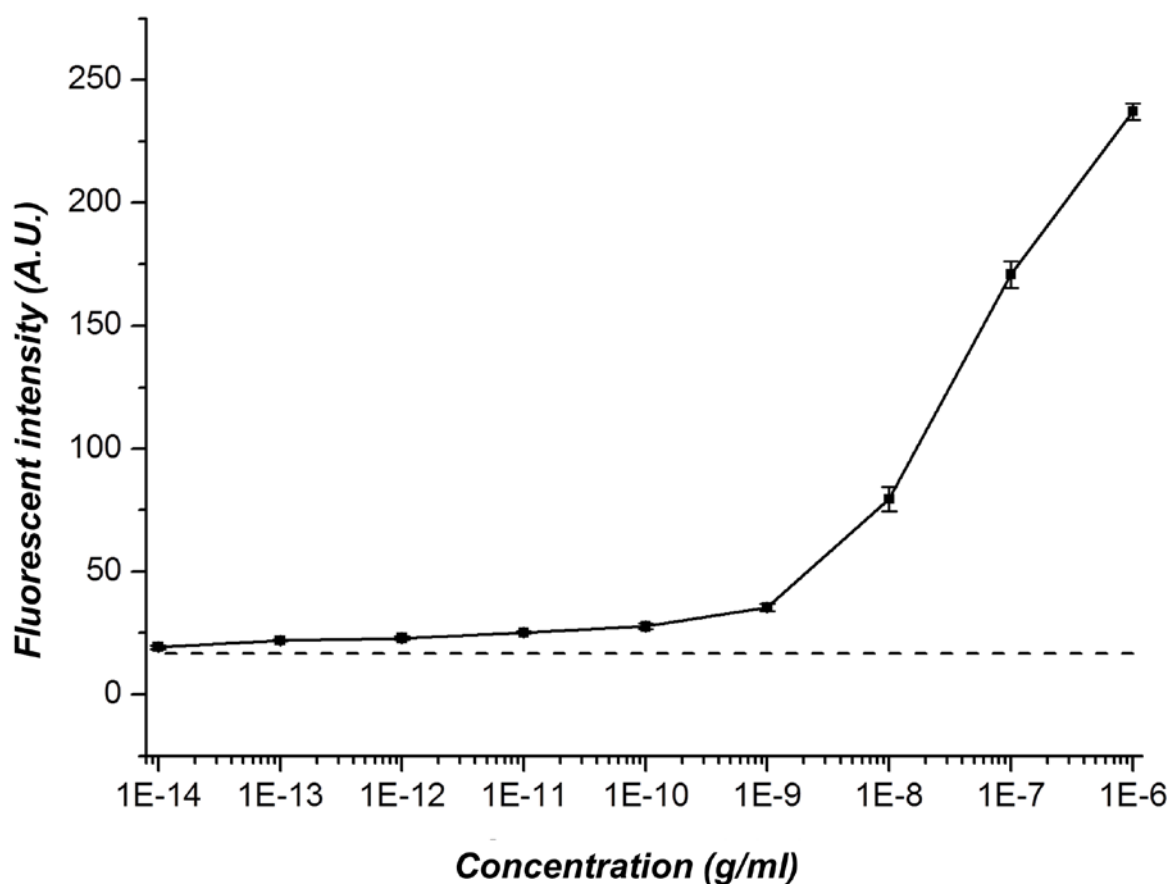
Fig. S5 **Detection sensitivity of hydrogel microfiber for biomolecular analysis.** Fluorescent intensity was measured corresponding to the concentration of FITC-anti human IgG. The distinguishable fluorescent signal of the hydrogel microfiber was as low as 0.01 pg/ml. The dotted line refers to fluorescent signal from the microfiber without treatment of FITC-anti human IgG.