Supporting Information

Microfluidic-driven short peptides hydrogels with optical waveguiding property

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1. Formation of peptide hydrogels in microfluidic devices.

The formation of the peptide hydrogel occurs at the central channel of the Y-shape microfluidic device. The well-controlled supply of buffers 1 (pH 12) and 2 (pH 5.8) through different inlets, into the Y-shaped channel, enables due to the laminar flow a contact governed by molecular diffusion between these two solutions. The contact/mixing occurs at the interface of the two buffers streams, at the central channel. The diffusion of the molecules at the interface between the two buffer solutions triggers the formation of the fibre once pH is around 7.4 (as reported previously for this tripeptide hydrogel).[1] The fibre is not formed instantaneously once the two buffers are in contact, but after some time (something that also happens in bulk conditions). Thus, in overall: first, the two buffers flow along the central channel filling it completely, time at which the pumps are stopped. At that moment, there is not any presence of the fibre yet (even when checking it at the fluorescence microscope). Instead, the formation of the fibre starts after around 2 min since the flow has been stopped. And then, the fibre continues growing for around 1h 30 minutes. The obtained fibre remains adhered to the glass substrate. In these conditions, the optical properties of the formed fibres can be measured (after peeling off the PDMS) (see Section 5).

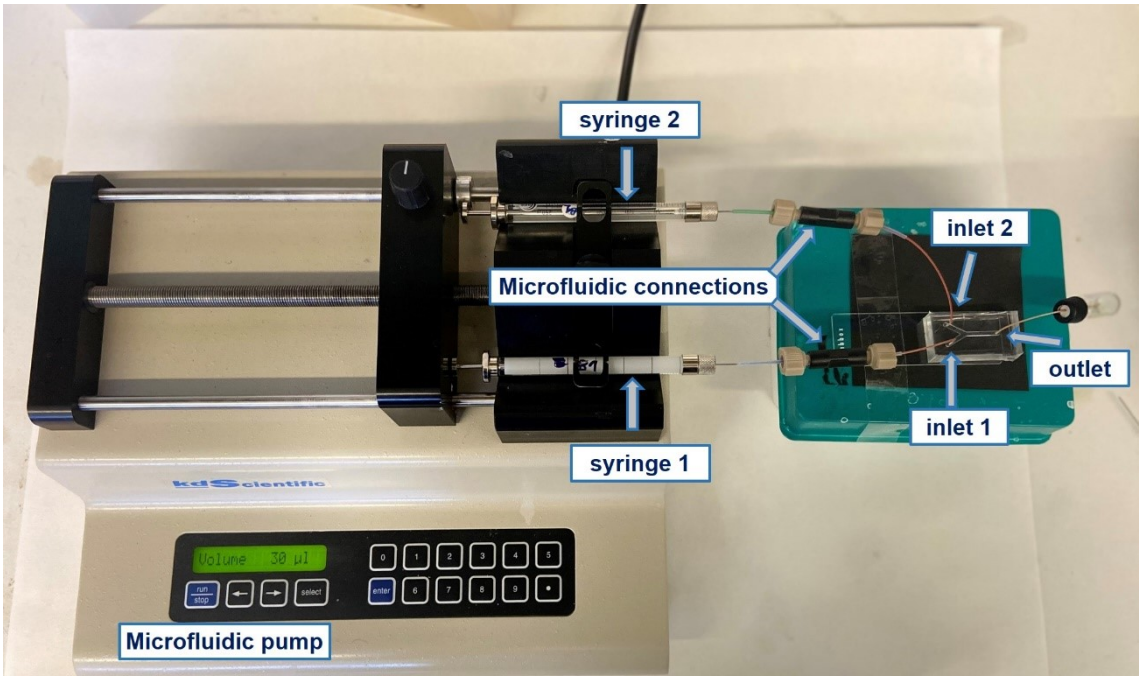

Figure S1. Photograph of the system used to prepare peptide hydrogel fibers.

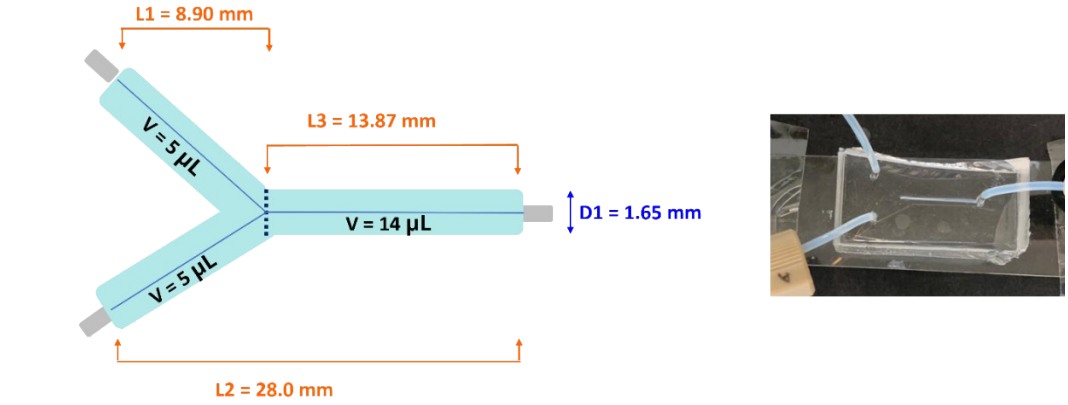

Figure S2. Scheme of the microfluidic devices used in this work, indicating the length and diameter of the channels, and their volume (left). Picture of the peptide hydrogel fiber formed in the absence of dyes, in a microfluidic device represented on the left (right).

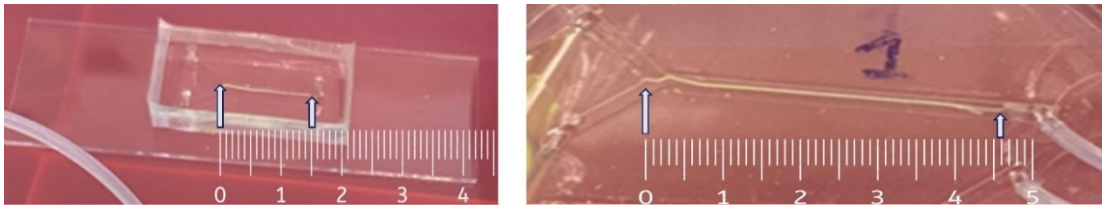

Figure S3. Picture of the peptide hydrogel fibre formed in microfluidic devices where the central channel has different lengths (1.5 cm and 4.5 cm). They are formed in the presence of ThT as dopant (peptide/dye molar ratio 102).

2. Fluorescence microscopy

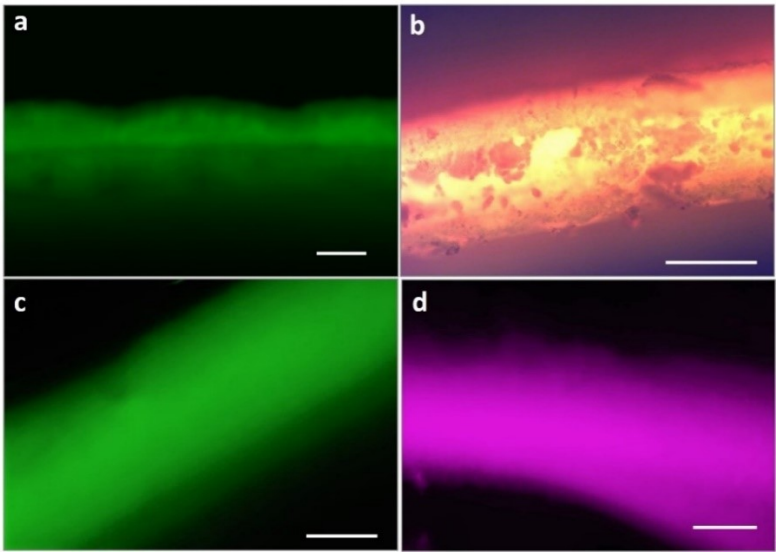

Figure S4. Fluorescence microscopy images of hydrogel fibers formed in the presence of ThT stock solution at 1 mg/mL (a,c) and RhB solution stock 0.5 mg/mL (b,d), using peptide concentration 8 mM (a,b) and 16 mM (c,d). Flow rate of 1 µL/min. Scale bar = 100 µm.

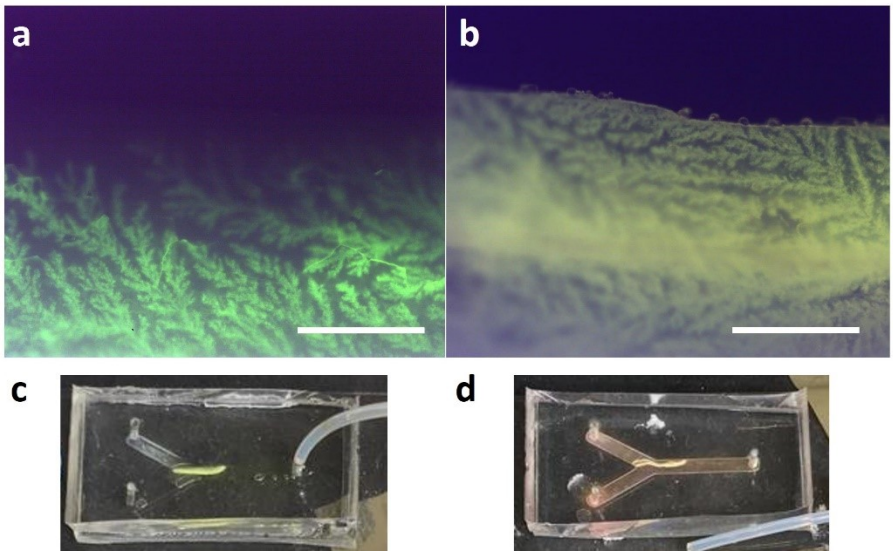

Figure S5. Fluorescence microscopy images (a,b) and photographs of devices (c,d) of hydrogel fibers at peptide concentration 16 mM in the presence of ThT stock at 1 mg/mL at flow rates of 0.3 μL/min (a) and 0.5 μL/min (b). Scale bar = 100 μm.

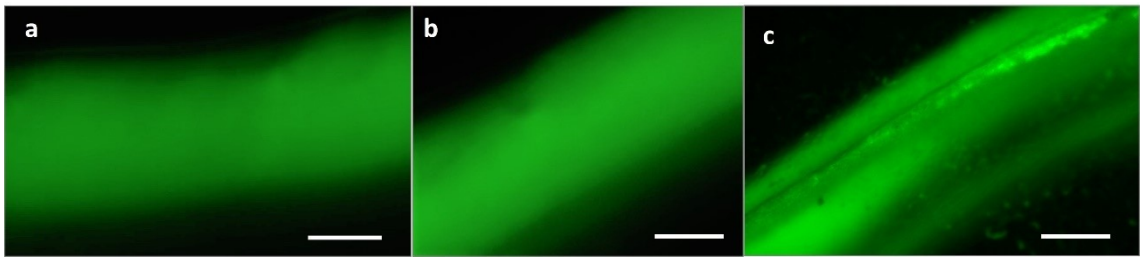

Figure S6. Fluorescence microscopy image of hydrogel fibers at peptide concentration 16 mM in the presence of ThT stock solution at 2 mg/mL (a), 1 mg/mL (b) and 0.5 mg/mL (c) (Flow rate: 1 μL/min). Scale bar = 100 μm.

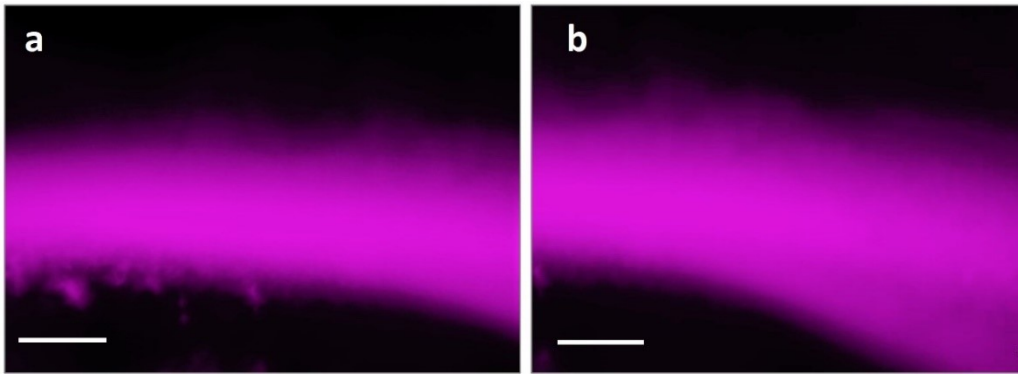

Figure S7. Fluorescence microscopy image of hydrogel fibers at peptide concentration 16 mM in the presence of RhB stock solution at 0.25 mg/mL (a) and 0.5 mg/mL (b) (Flow rate: 1 μL/min). Scale bar = 100 μm.

3. Raman spectroscopy

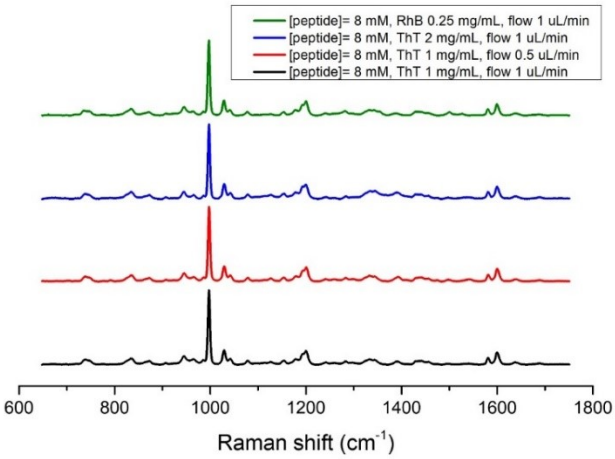

Figure S8. Raman spectra of hydrogel fibers formed in the presence of ThT and RhB, varying dye concentration and flow rate.

4. Scanning electron microscopy

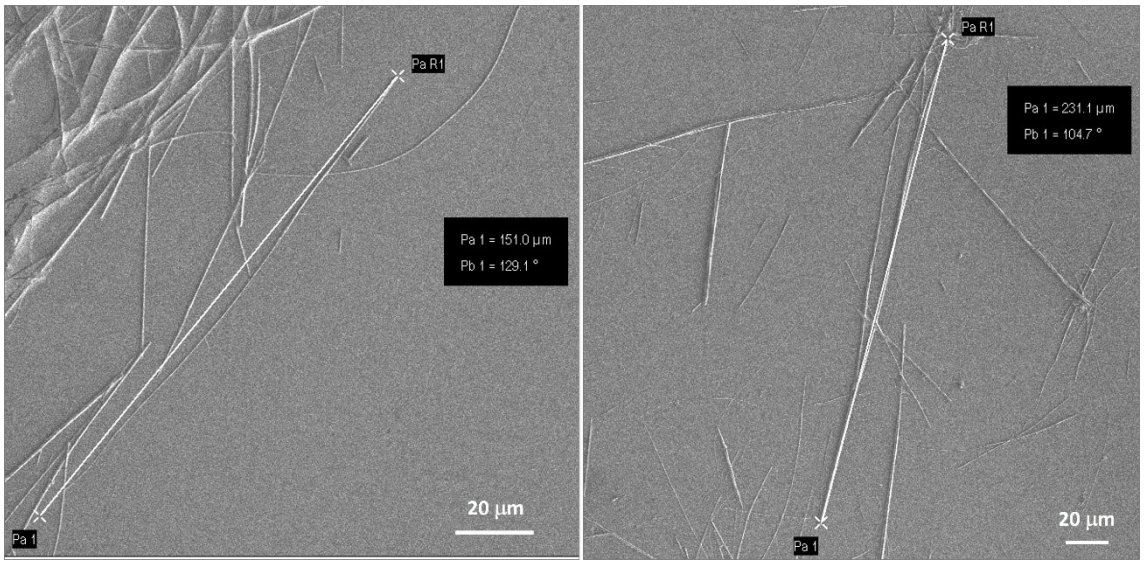

Figure S9. SEM images of peptide hydrogel prepared in bulk. It can be clearly seen that orientation is random and not well-controlled unlike in the case of microfluidics-assisted hydrogel formation.

5. Optical waveguide measurements

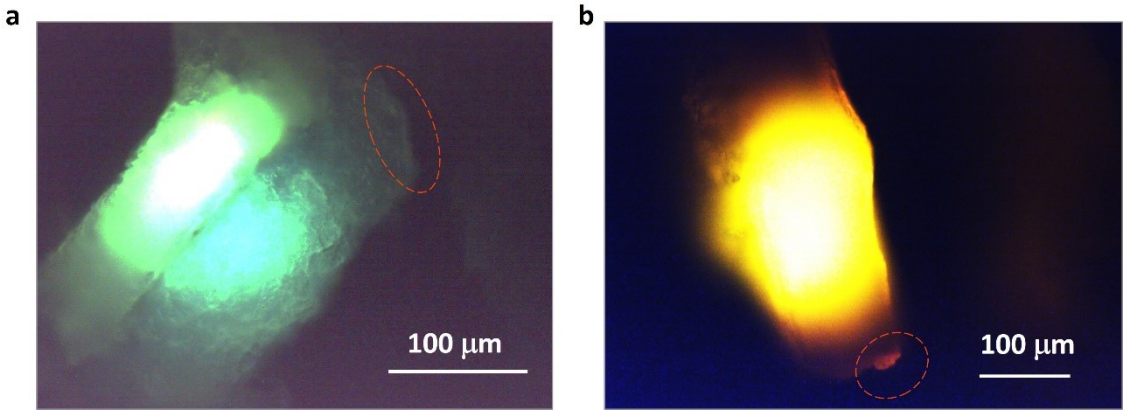

Figure S10. PL microscopy images of ^{D}Leu-^{L}Phe-^{L}Phe hydrogels prepared in microfluidics using peptide solution at 16 mM concentration in the presence of (a) ThT (stock 1 mg/mL, peptide/ThT molar ratio 102) and (b) RhB (stock 0.5 mg/mL, peptide/RhB molar ratio 305), respectively (Flow rate: 1 μL/min). The red circles indicate the exit of fluorescence light.

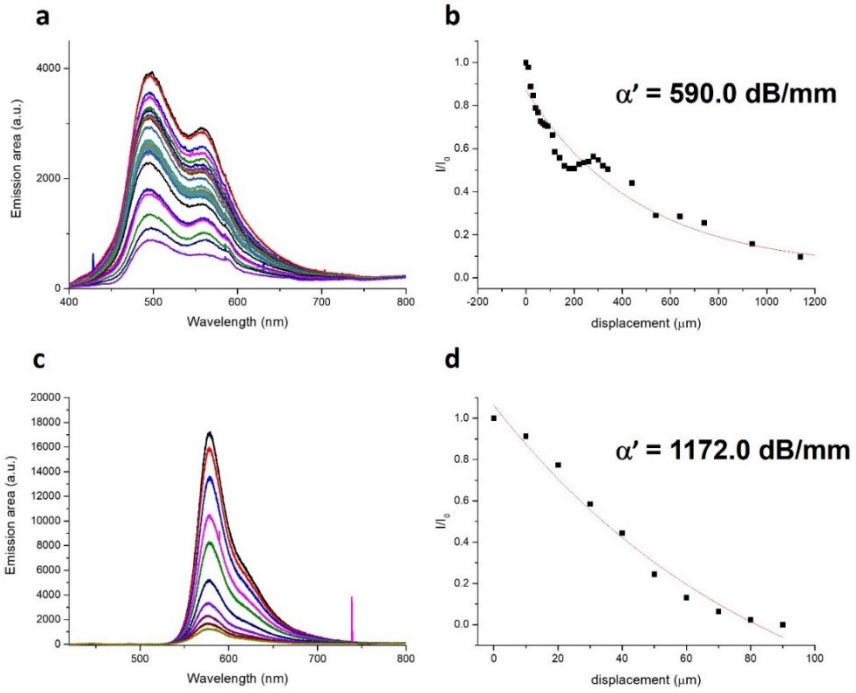

Figure S11. Fluorescence spectra (a,c) and optical loss coefficient calculation (b,d) for hydrogel fibers at 8 mM of peptide concentration with ThT stock 1 mg/mL (a,b) and RhB stock 0.5 mg/mL (c,d) (Flow rate: 1 μL/min).

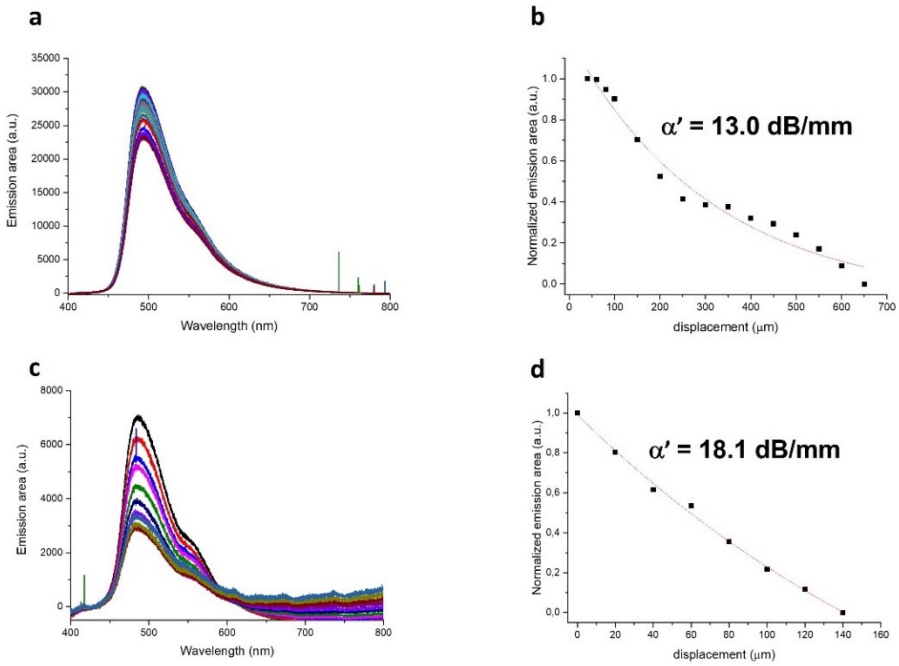

Figure S12. Fluorescence spectra (a,c) and optical loss coefficient calculation (b,d) for hydrogel fibers at peptide concentration 16 mM in the presence of ThT at 2 mg/mL (a,c) and 0.5 mg/mL (b,d) stock solution concentration. (Flow rate: 1 μL/min).

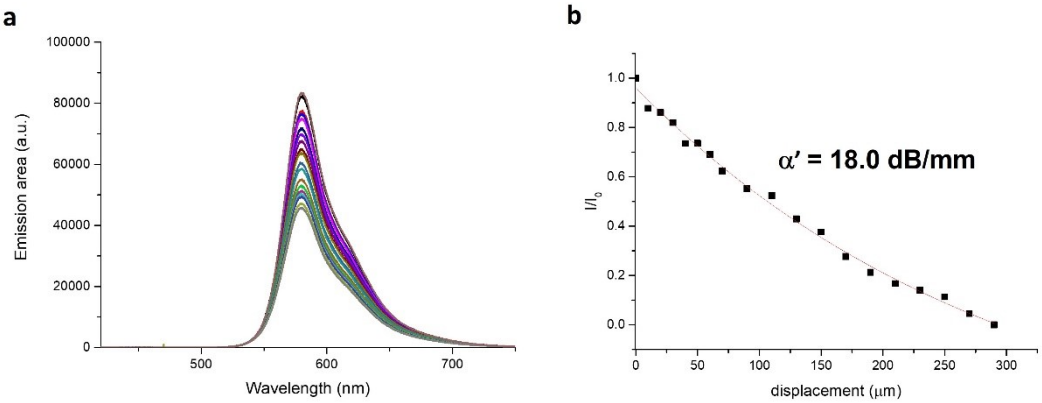

Figure S13. Fluorescence spectra (a) and optical loss coefficient calculation (b) for hydrogel fiber at 16 mM with RhB stock 0.25 mg/mL (Flow rate: 1 μL/min).

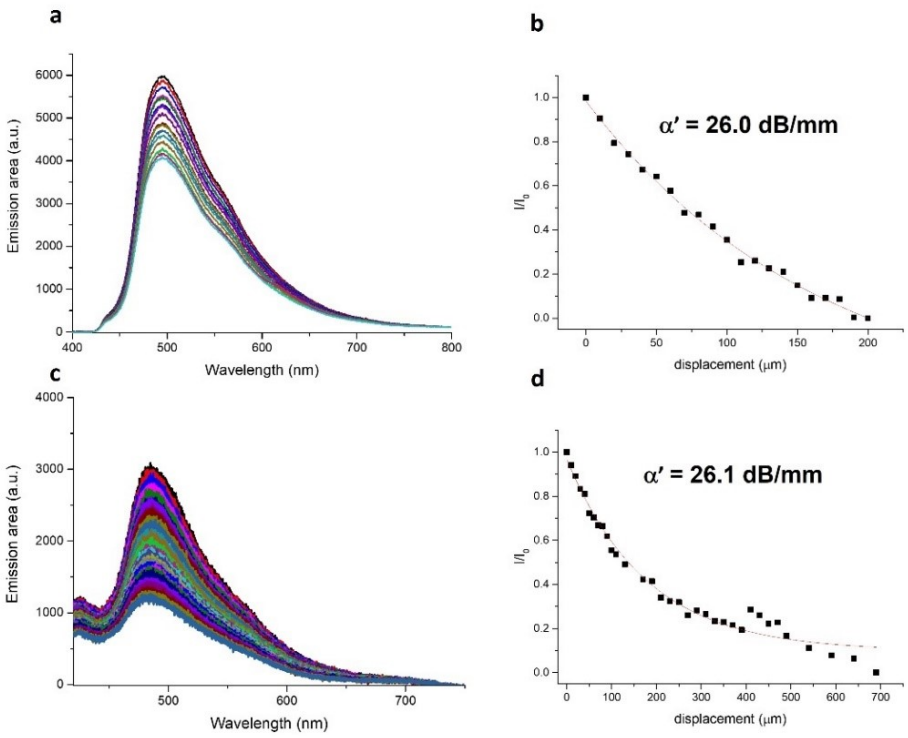

Figure S14. Fluorescence spectra (a,c) and optical loss coefficient calculation (b,d) for hydrogel fibers at peptide concentration 16 mM in the presence of ThT stock solution at 1 mg/mL at flow rates of 0.5 μL/min (a,b) and 0.3 μL/min (b,c).

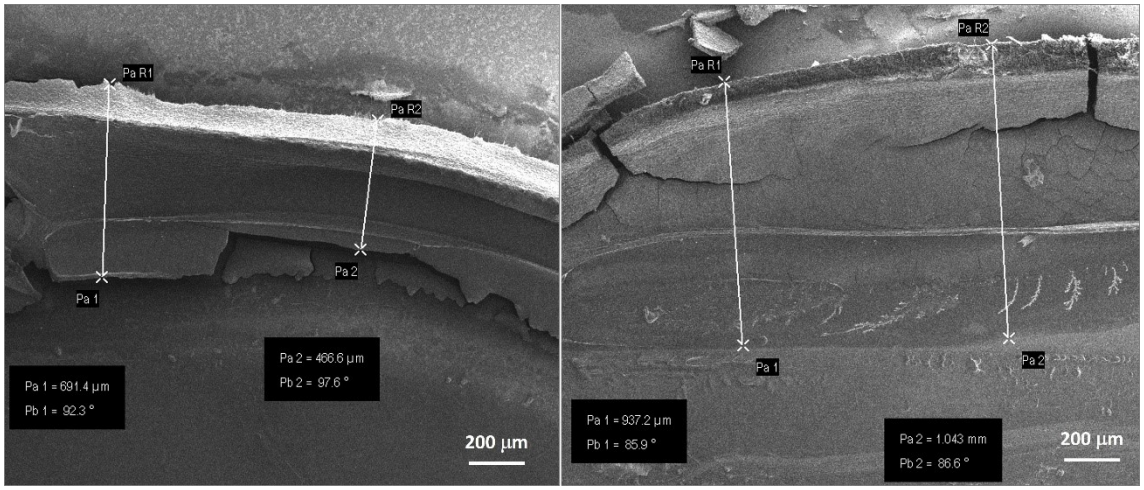

Figure S15. SEM images of peptide hydrogel prepared in microfluidic devices at different flow rates: (a) 0.5 μL/min and (b) 0.3 μL/min (rest of conditions are the optimum ones: peptide concentration 16 mM, ThT stock solution 1 mg/mL). It can be clearly seen that these fibers are thicker and present more structural defects than those formed at a flow rate 1 μL/min.