Supporting Information

Biomimetic Self-Templated Hierarchical Structures of Collagen-like Peptide Amphiphiles

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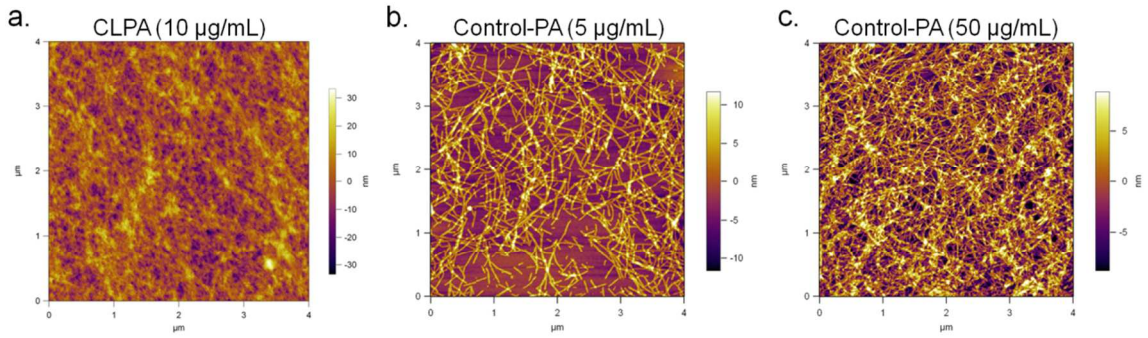


Figure S1. AFM images of drop cast CLPA and Control-PA on Si substrates. **a-c**, 10 µL of solution was dropped on a Si wafer and dried at room temperature.

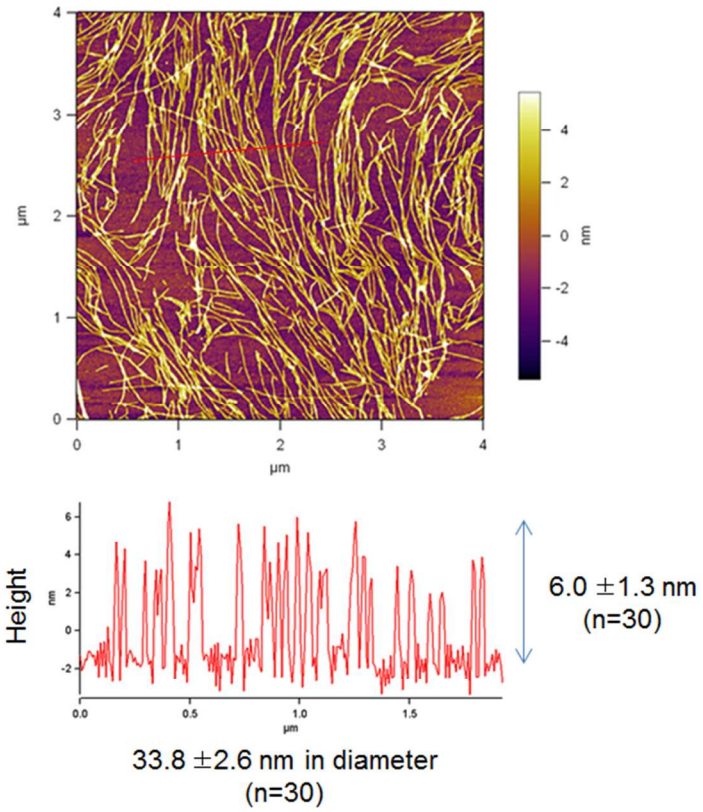


Figure S2. AFM image and cross-sectional profile of CLPA nanofibers. The nanofibers were formed from a drop-cast CLPA solution (10 μg/mL).

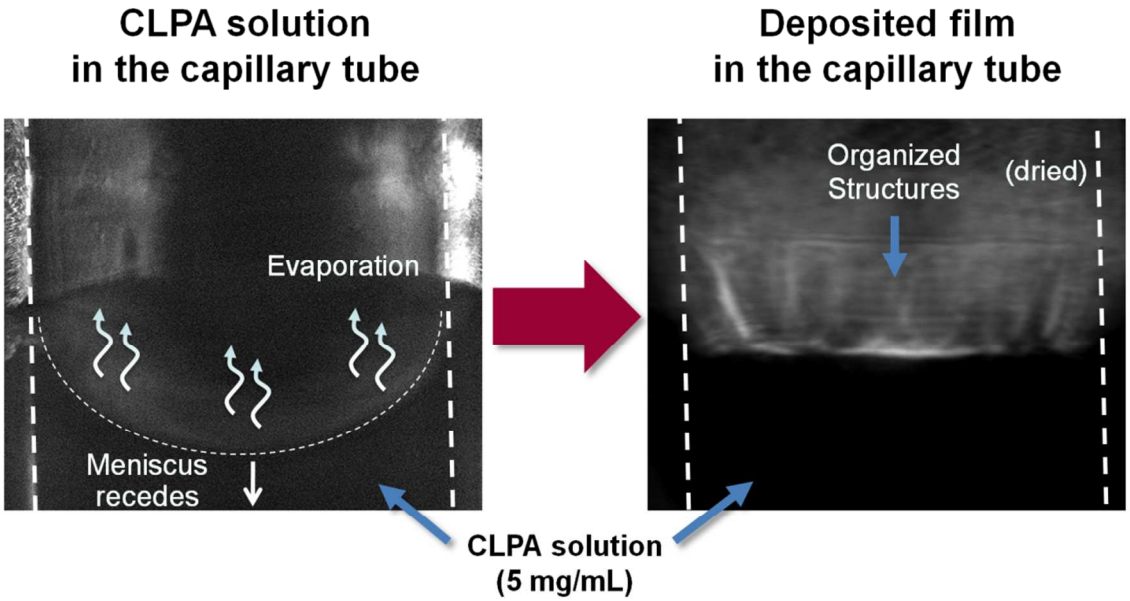


Figure S3. Polarized optical microscope image of the meniscus area of CLPA (5 mg/mL) filled in the capillary tube. As the meniscus receded the organized structures were deposited on the wall of the capillary tube.

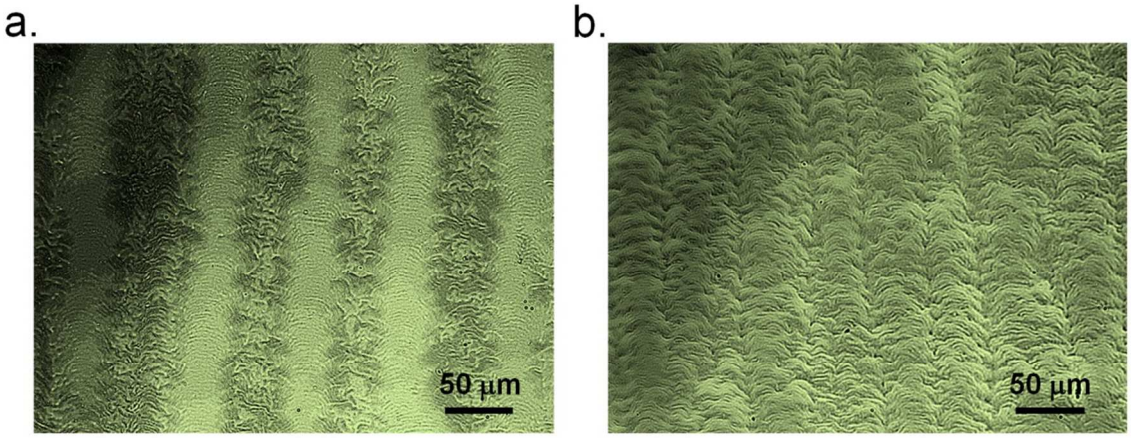


Figure S4. Optical microscope images of twisted plywood-like structures created by pulling a substrate at: 30 μm/min (a); and 70 μm/min (b) from 1 mg/mL of CLPA solution.

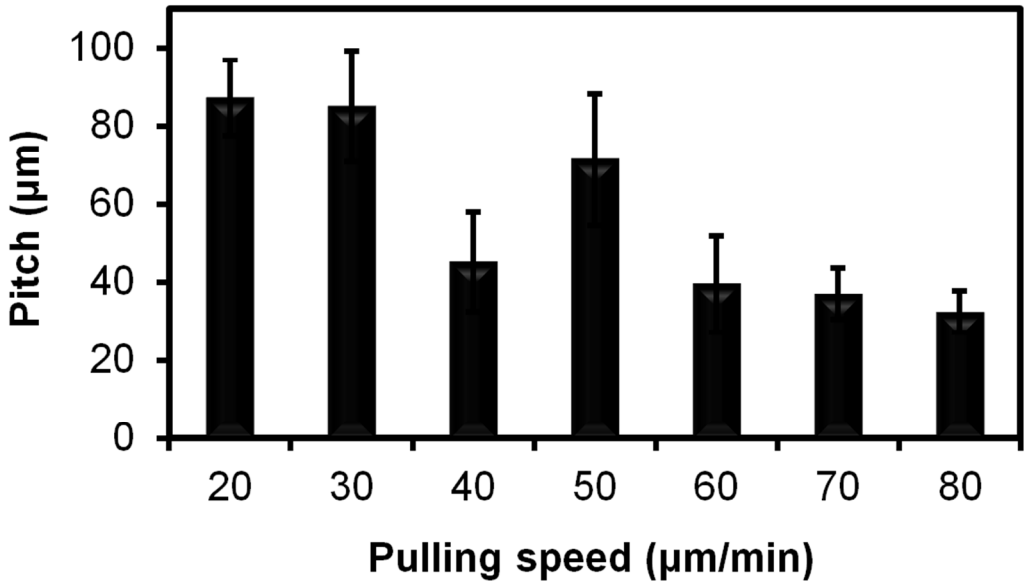


Figure S5. Structural periodicity in twisted plywood-like phase structures (1 mg/mL) depends on pulling speed.

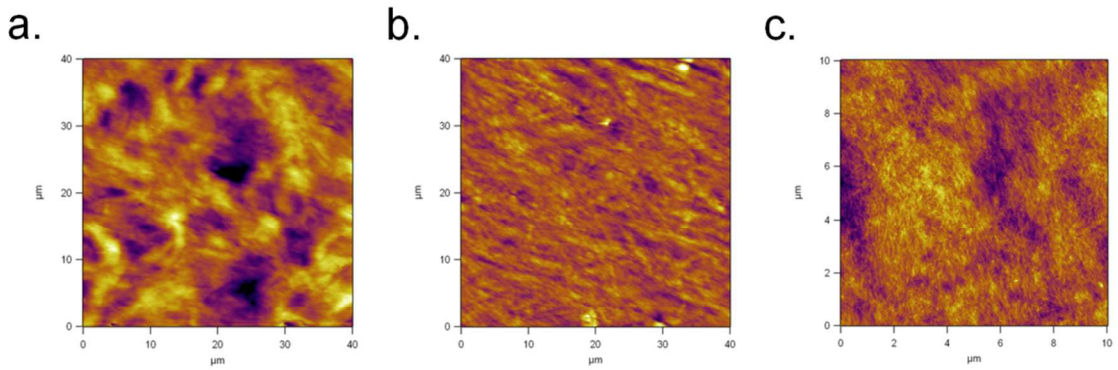


Figure S6. AFM images of the self-templated Control-PA structures. **a**, pulled at 30 μm/min from 10 mg/mL of Control-PA solution. **b**, pulled at 50 μm/min from 10 mg/mL of Control-PA solution. **c**, pulled at 30 μm/min from 5 mg/mL of Control-PA solution.

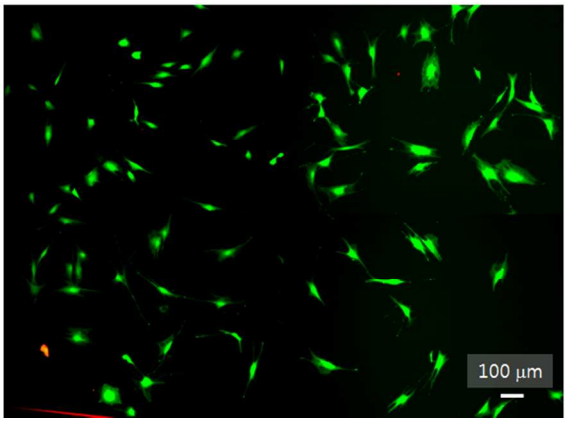


Figure S7. Cell viability on the drop-casted CLPA film. MC-3T3 E1 cells were used at passage number 12 and seeded at a density of ~2 x 10^2 cells/cm^2 on top of the CLPA films (drop-cast). The viability of cells grown on the film for five days was measured using Live/Dead viability kit (Invitrogen, Carlsbad, CA) according to the manufacturer's instruction (live cells: green, dead cells: red). The CLPA supported over 95% cell viability throughout the experiment.

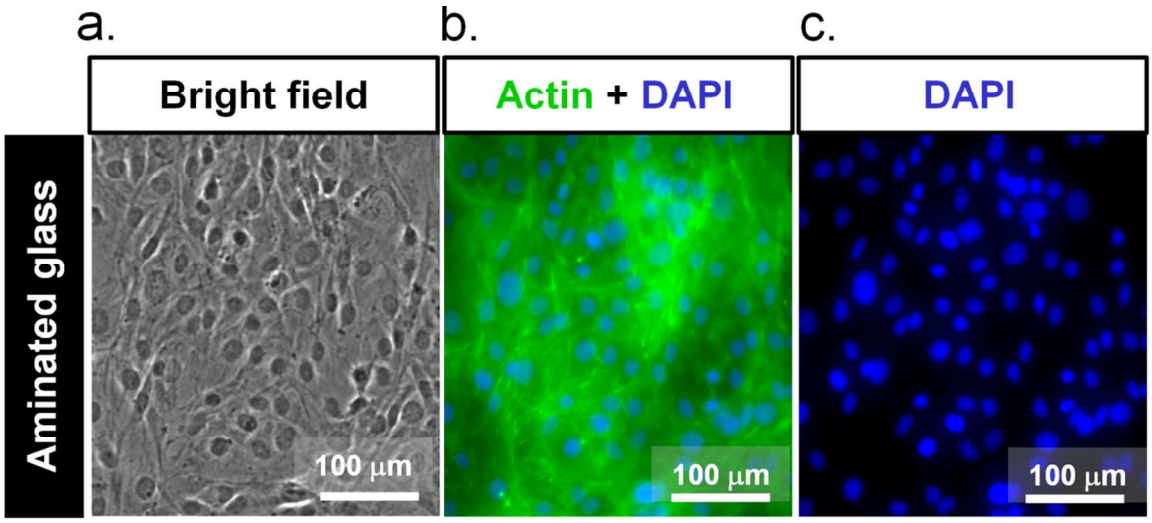
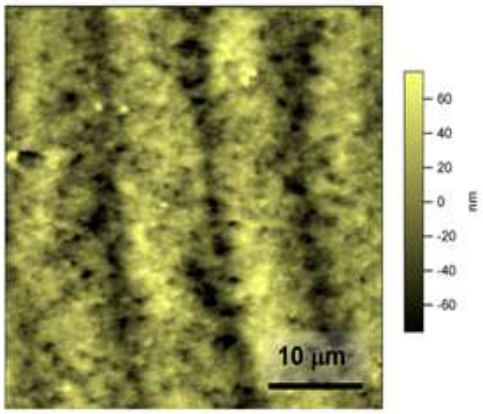
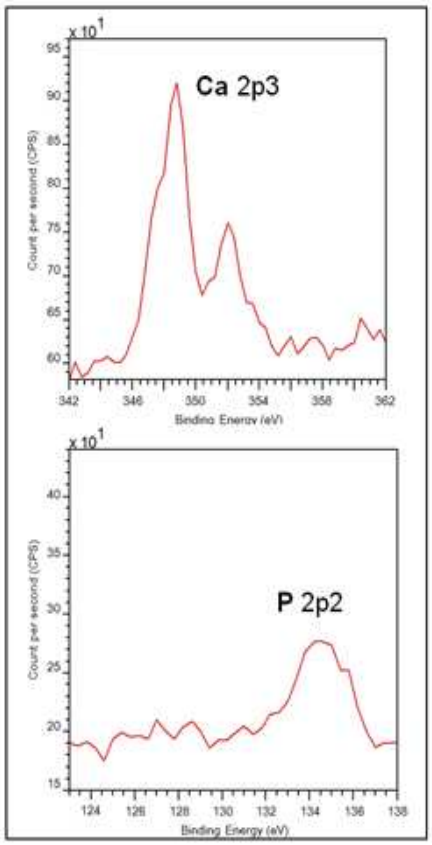


Figure S8. Microscope images of MC-3T3 cells cultured (five days) on glass (pre-treated with 3-aminopropyltriethoxysilane). (A) bright field, (B) merged fluorescence (actin:green, nuclei:blue) and (C) nuclei. The cells grew without preferred orientation on the glass substrates.

a.



b.



c.

No.	Atomic ratio (Ca : P)
1	1.70 : 1
2	1.65 : 1
3	1.56 : 1
Avg.	**1.64 : 1**

Figure S9. Self-templated CLPA mineralized by alternate dipping into Ca^{2+} (100 mM) and PO_4^{3-} (100 mM) solution 50 times. Between each dipping the film was rinsed with DI water. **a**, AFM image of the mineralized film showing the preserved micro-structural order. **b & c**, XPS analysis of the mineralized CLPA film showed a calcium to phosphorous atomic ratio of 1.64:1.

Table S1. Comparison of peptide amphiphile and M13 phage in self-templating assembly

Features		Peptide amphiphile (PA)	M13 phage[1]
Molecular design	Site-specific multi-functional peptide display	Not available	Display on the shaft of the phage body (pVIII) and at either tips (pIII, pVII, pIX)
	Displayable peptide with high density	- Net charge: (+), (-) (CLPA has (+) net charge) - Length: commonly 6-20 amino acids - Amino acids (AAs): natural, unnatural and synthetic - Crosslinkable AA: multiple cysteines	- Net charge: (-) is favorable on pVIII - Length: up to 8-10 amino acids on pVIII - Amino acids (AAs): only natural - Crosslinkable AA: cysteine is disfavored on pVIII
Building block production	Method	- Chemical method - Use of organic solvents in synthesis - Non-amplifiable	- Biological method - Use of water-based media in production - Amplifiable (via E. coli infection)
	Yield	~300 mg in a lab scale	~30 mg in a lab scale
Assembly property	Liquid crystalline behavior	- PA structure-LC behavior relationship has not been fully explored yet - Concentration-dependent	- Well studied and established - Concentration-dependent (nematic, cholesteric, smectic, chiral smectic)
	Self-templated structures	In CLPA case, - Twisted plywood-like phase (1 mg/mL) - Crimped filament-phase (3-5 mg/mL)	- Nematic orthogonal twists and cholesteric helical ribbons (1-2.0 mg/mL) - Smectic helicoidal filaments (3-6 mg/mL)

Reference

1. Chung W-J*, et al.* Biomimetic self-templating supramolecular structures. *Nature* **478**, 364-368 (2011).