

## Supporting Information

# **Bio-Inspired Silkworm 3D Cocoon-like Hierarchical Self-assembled Structure from $\pi$ -conjugated Natural Aromatic Amino Acids**

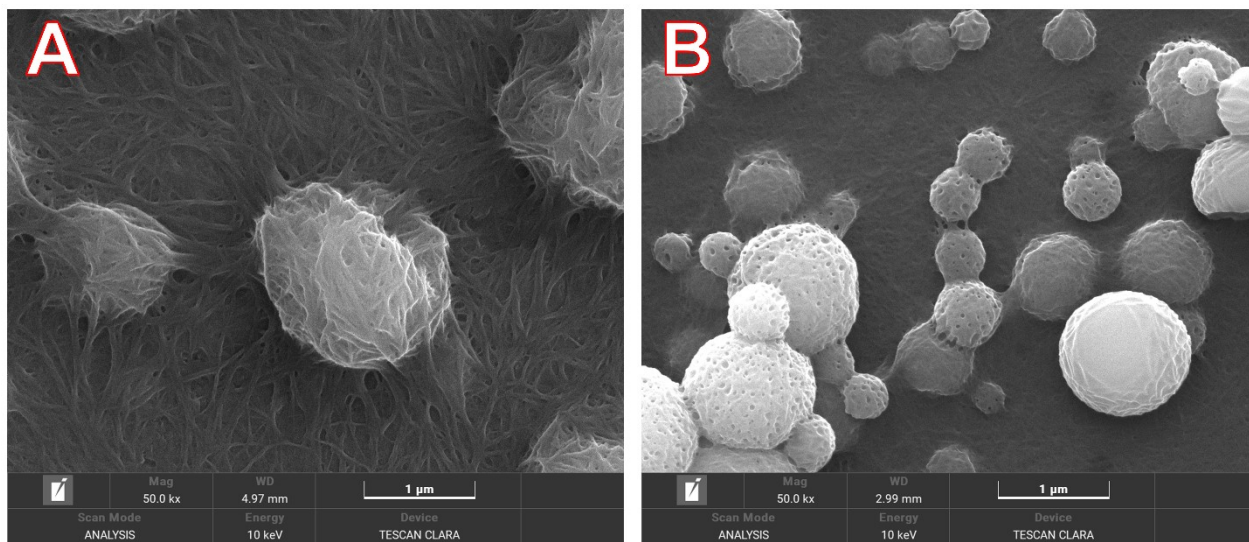
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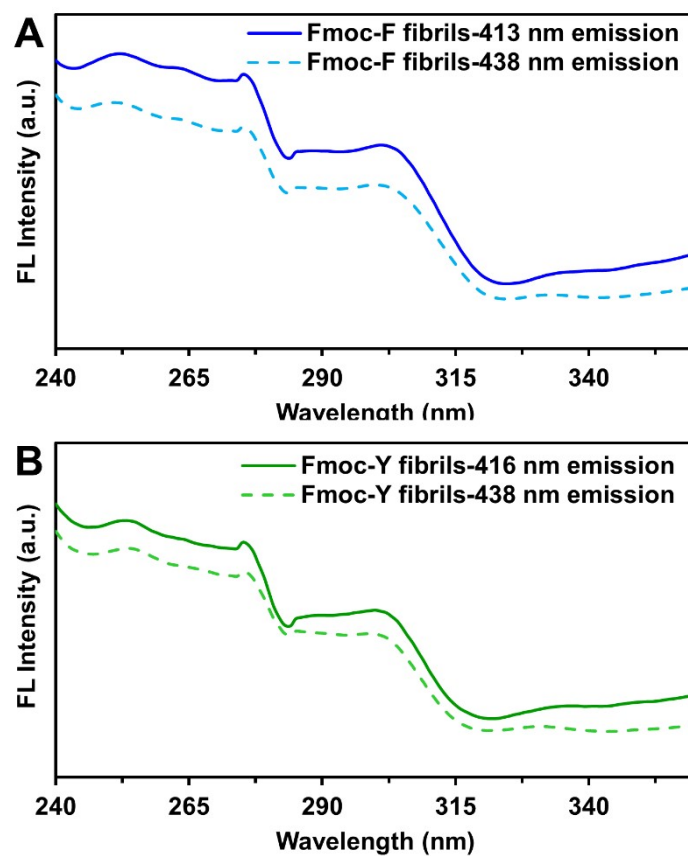
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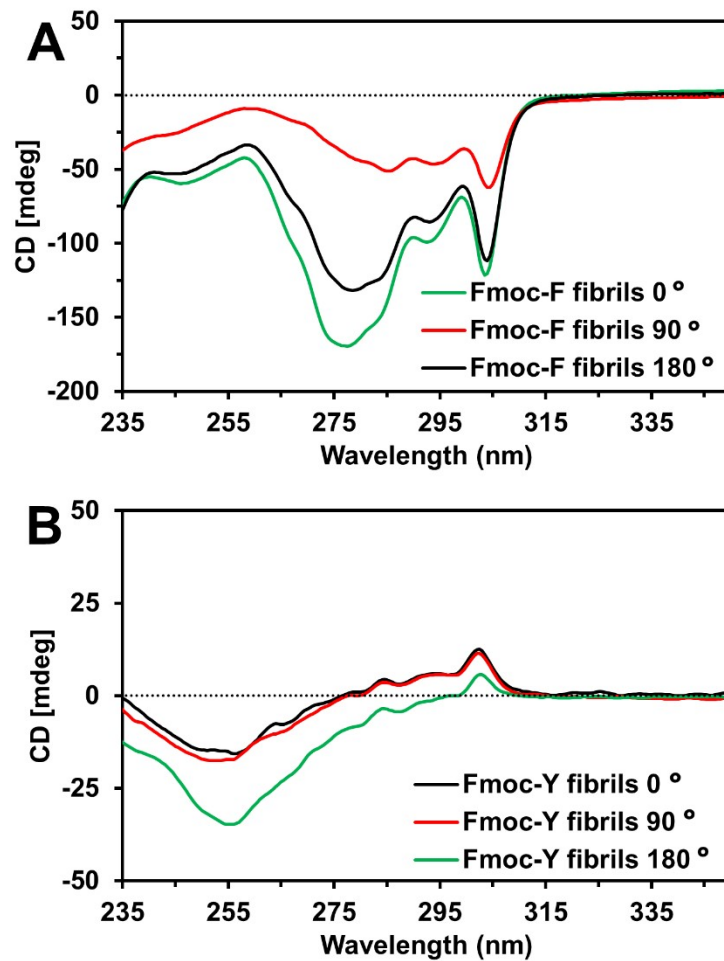
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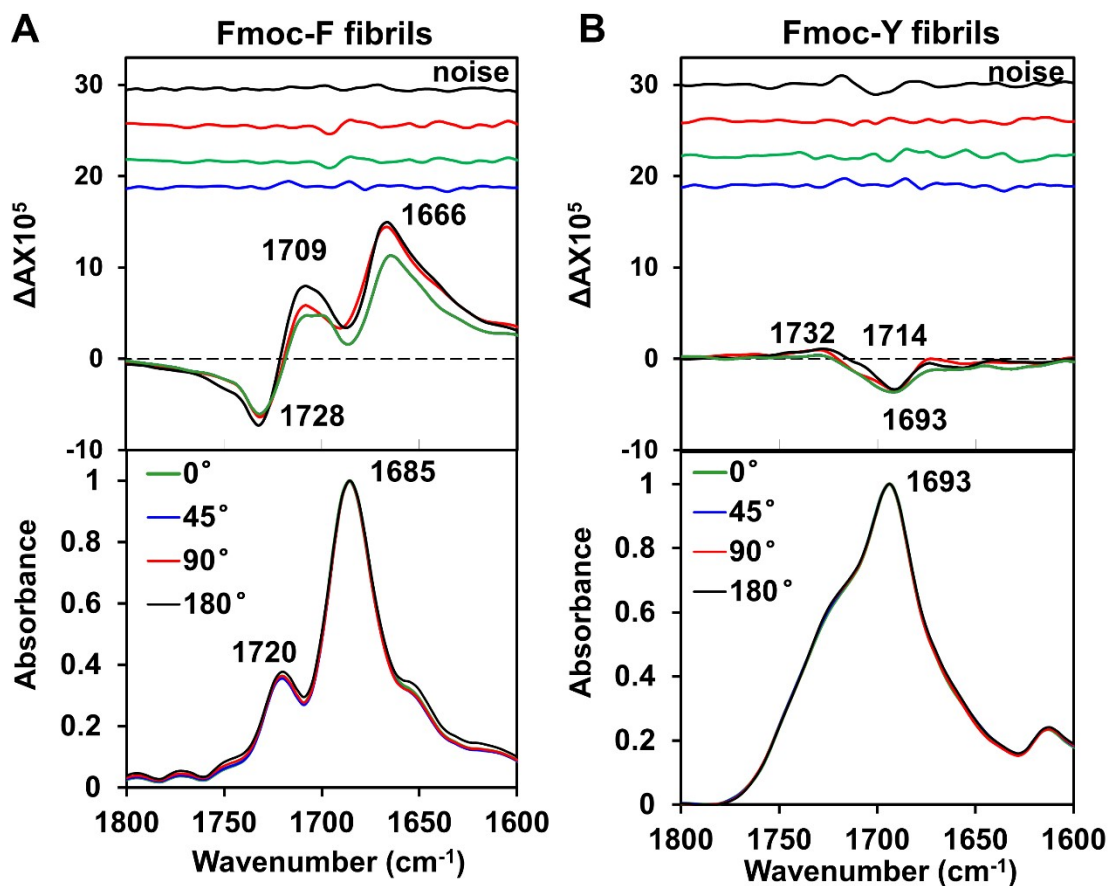
**Figure S1.** Additional FE-SEM images of (A) Fmoc-F and (B) Fmoc-Y cocoon-like structures formed on drying at 30 °C and 27% RH. The cocoon-like structures are composed of entangled fibrils.



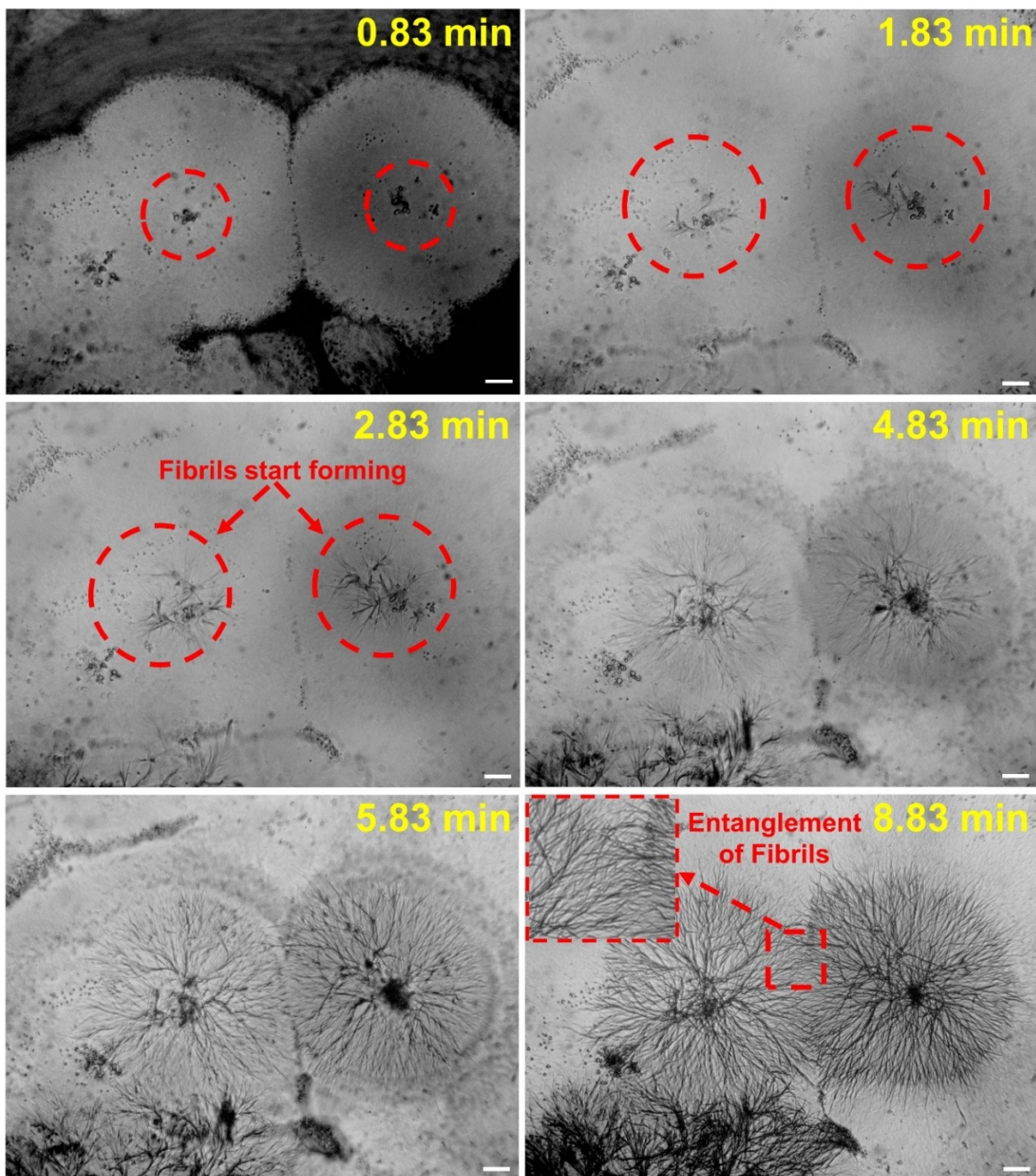
**Figure S2.** Excitation spectra of (A) Fmoc-F and (B) Fmoc-Y fibrils at two emission wavelengths.



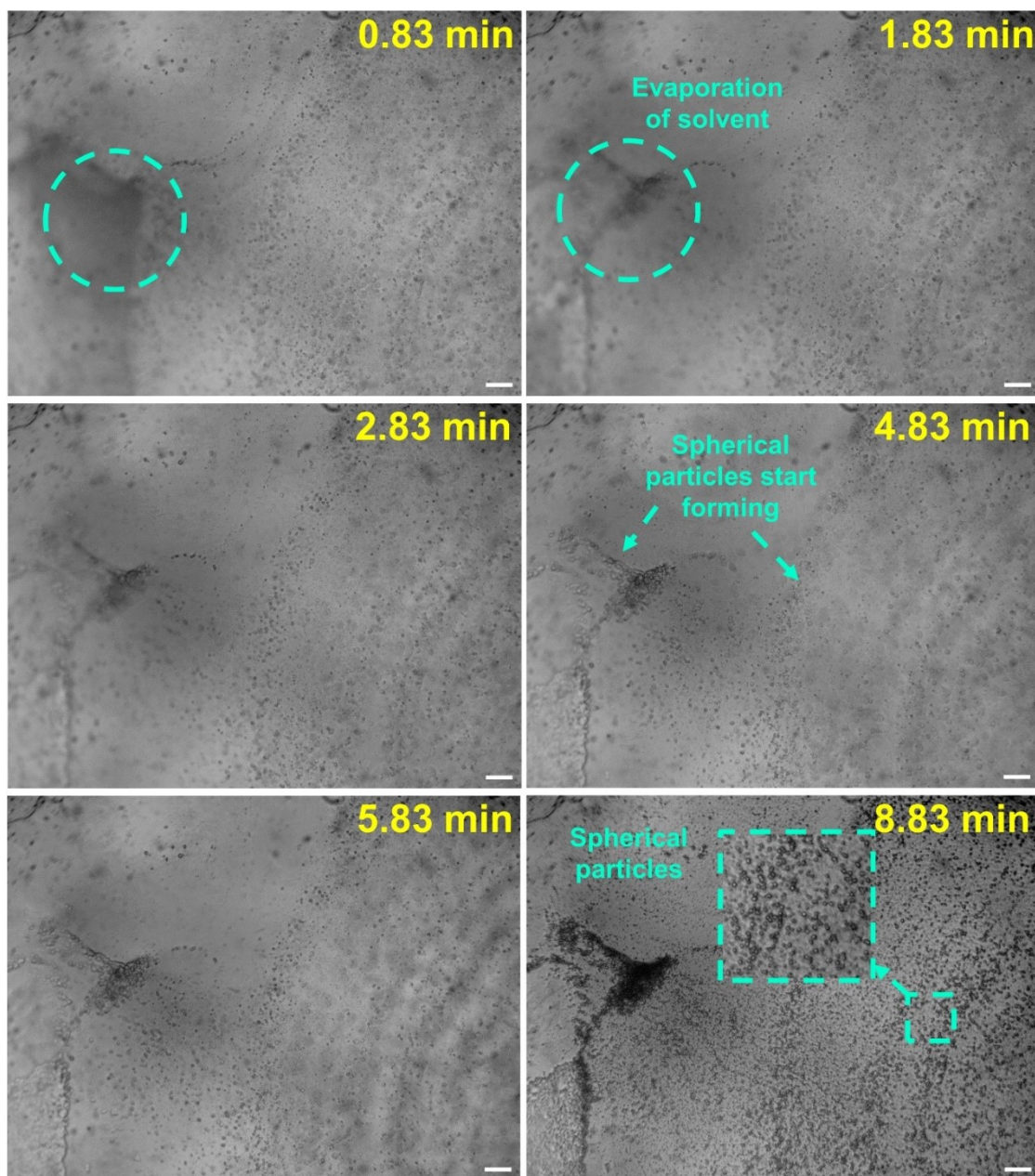
**Figure S3.** ECD spectra of (A) Fmoc-F and (B) Fmoc-Y fibrils, formed on the quartz slide, at different angles (0°, 90° and 180°) with respect to the light beam axis.



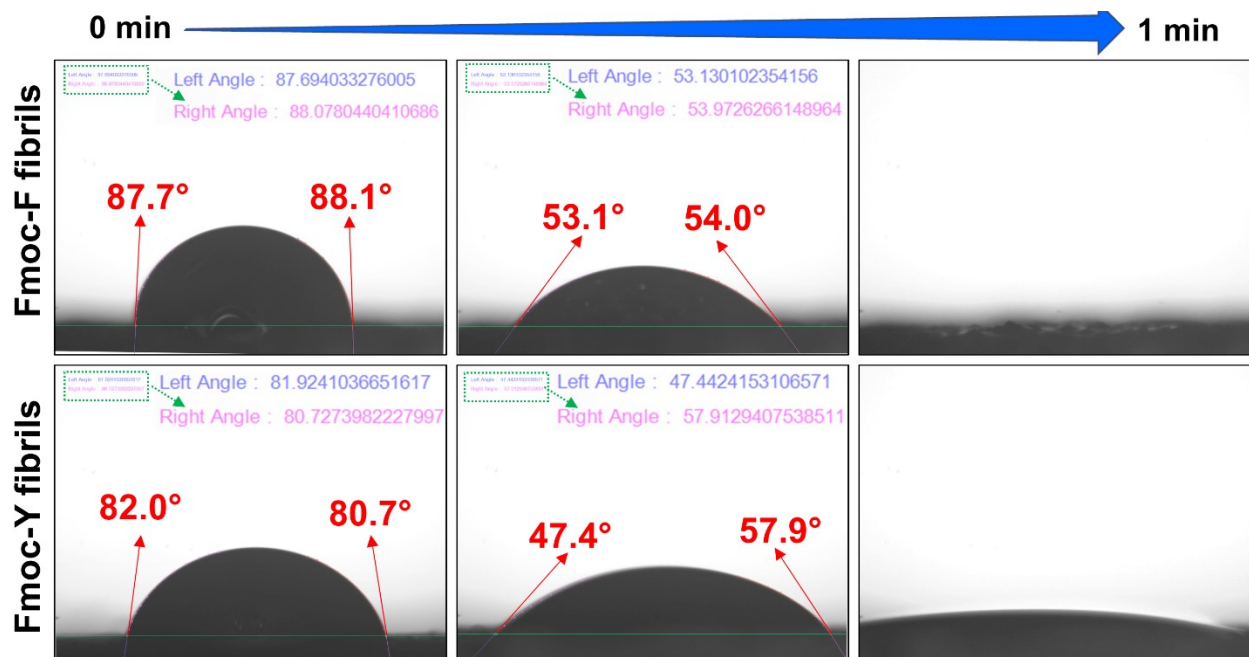
**Figure S4.** Vibrational absorption (bottom panel) and VCD (top panel) spectra of (A) Fmoc-F and (B) Fmoc-Y fibrils (supported on CaF<sub>2</sub> plate) obtained from different rotations ( 0°, 45°, 90° and 180° ) around the light beam axis.



**Figure S5.** Time-lapse optical microscopic images (40X) of Fmoc-F fibrils forming during drying from a 2 mg/mL solution in ethanol (Scale bar = 10  $\mu\text{m}$ ). The rate of evaporation is slower here than in confocal laser scanning microscopy since the optical microscopy experiment was conducted in a closed environment with less air circulation. It is note that the cocoon-like structures of diameter about 1  $\mu\text{m}$  could not be visualised using an optical microscope due to its own limitation.



**Figure S6.** Time-lapse optical microscopic images (40X) of Fmoc-Y fibrils forming during drying from a 2 mg/mL solution in ethanol (Scale bar = 10  $\mu\text{m}$ ). The rate of evaporation is slower here than in confocal laser scanning microscopy since the optical microscopy experiment was conducted in a closed environment with less air circulation. Due to the low resolution of optical microscope, the smaller diameter Fmoc-Y fibrils could not be visualized. It is note that the cocoon-like structures of diameter about 1  $\mu\text{m}$  could not be visualised using an optical microscope due to its own limitation.



**Figure S7.** Change in water contact angle on Fmoc-F and Fmoc-Y fibrils-coated glass surfaces as a function of time, demonstrating that water drops spread quickly on the coated glass surfaces. The left and right water contact angles are highlighted on the images in red.