Supporting Information for

Controlling Supramolecular Chiral Nanostructures by Self-Assembly of a Biomimetic β-Sheet-rich Amyloidogenic Peptide

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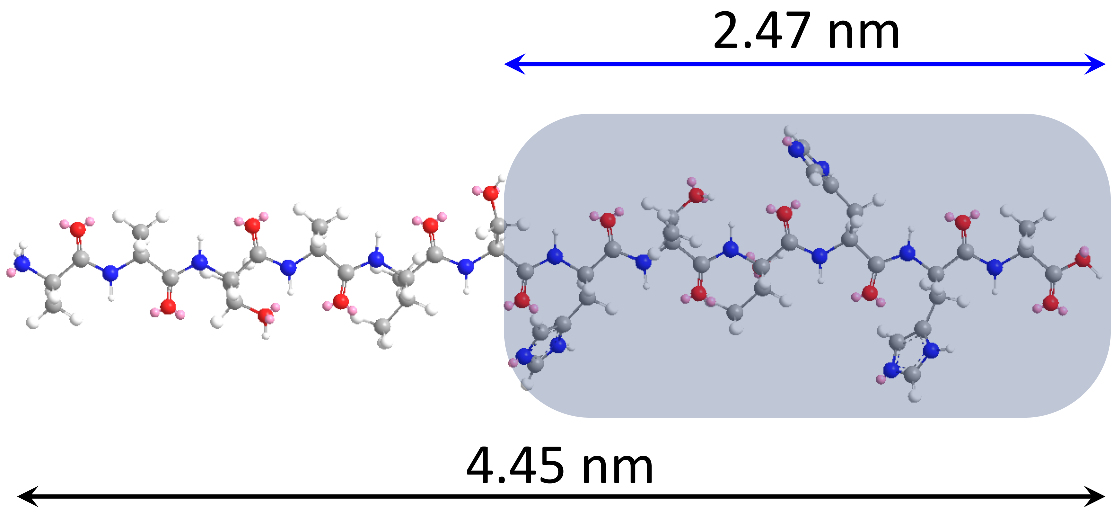


Figure SI-1. The chemical structure of **A1H1**, and the dimensions of the fully extended peptide sequence and the hydrophilic domain.

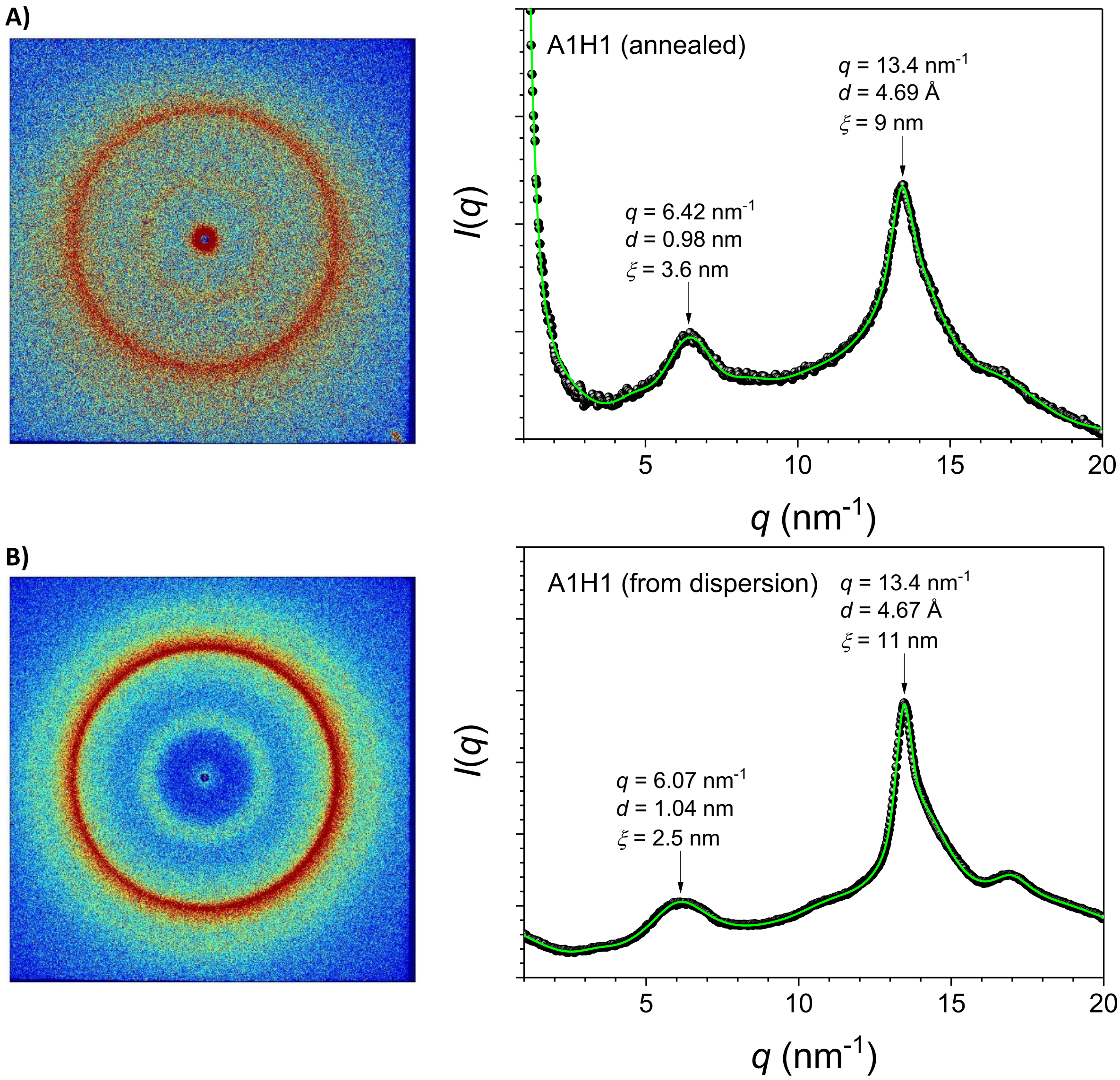


Figure SI-2. 2D and 1D WAXS intensity profile for **(A)** the annealed **A1H1** sample at 120 °C under nitrogen atmosphere, and **(B)** the dried **A1H1** sample from the 2 wt-% **A1H1** dispersion.

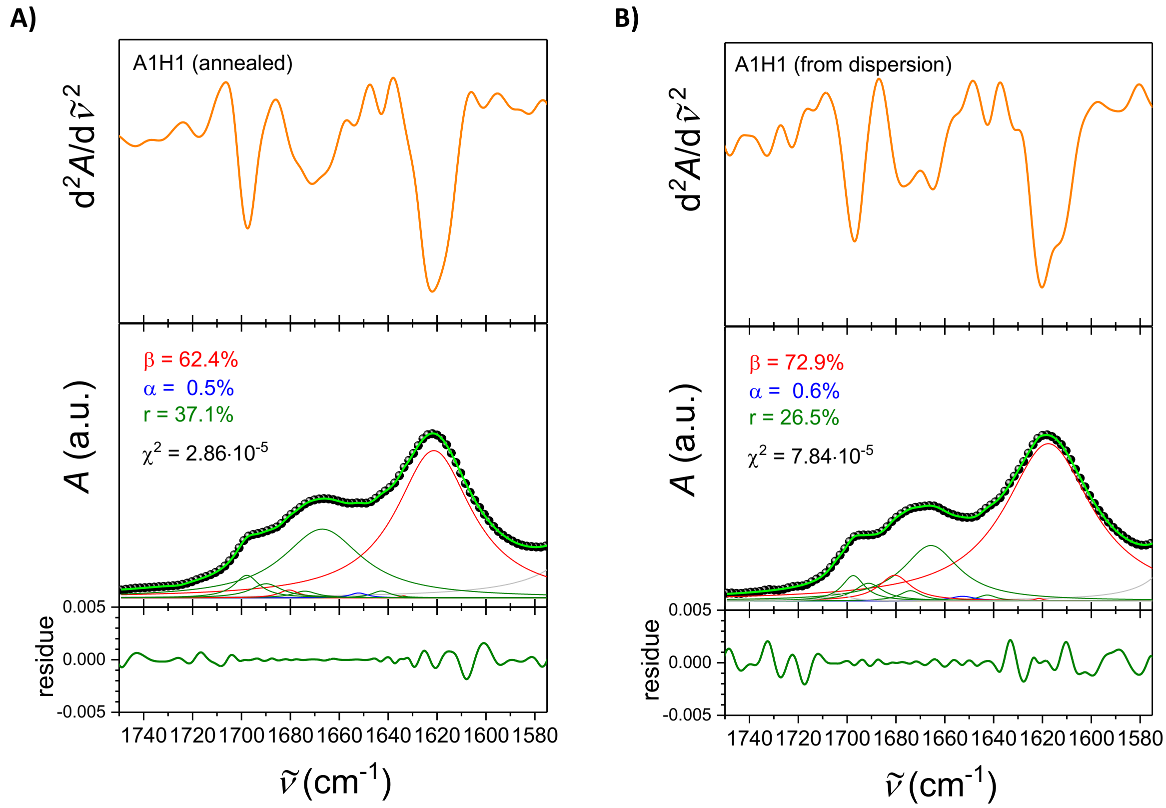
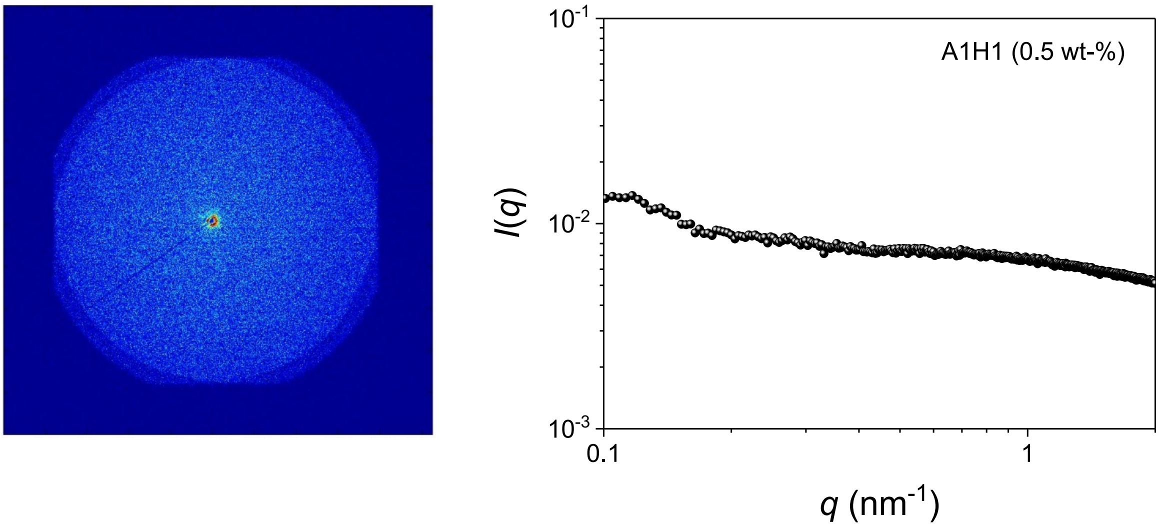
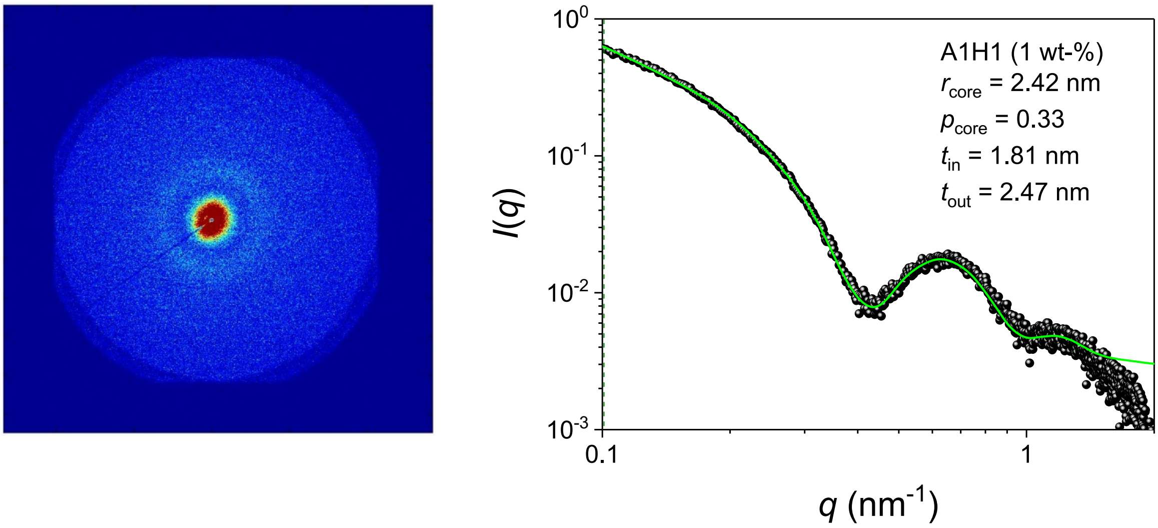


Figure SI-3. FTIR spectrum and peptide secondary structure population analysis for (**A)** the annealed **A1H1** sample at 120 °C under nitrogen atmosphere, and **(B)** the dried **A1H1** sample from the 2 wt-% dispersion. *Note*: β: β-sheets; α: α-helices; r: random coils.

A)



B)



C)

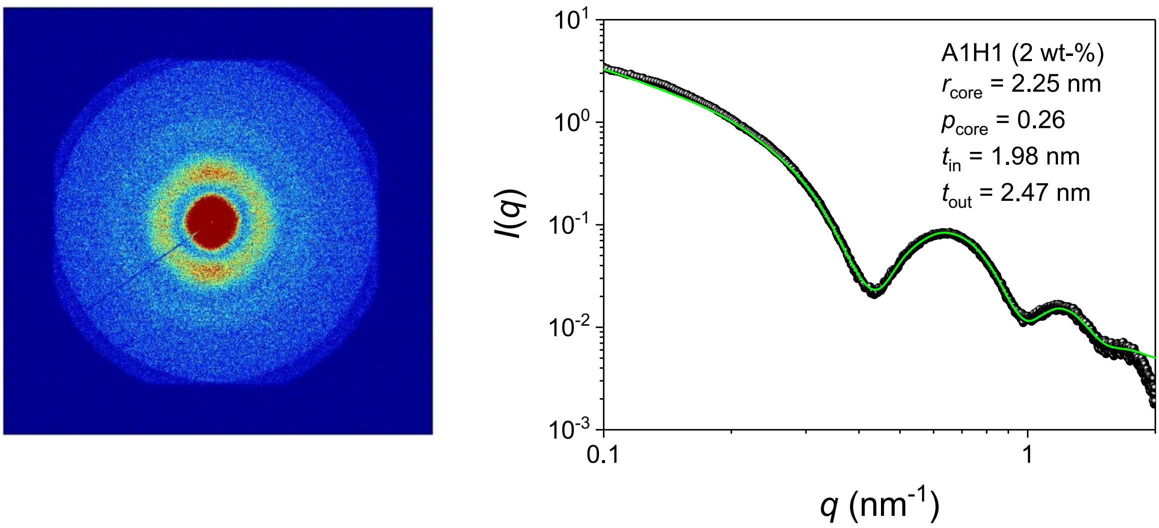


Figure SI-4. 2D and 1D SAXS intensity profile for **A1H1** dispersions in acetonitrile/water at **(A)** 0.5 wt-%, **(B)** 1 wt-%, and **(C)** 2 wt-%. *Notes:* capillaries were placed horizontally; the green fitting curve correspond to the form factor $P(q)$.

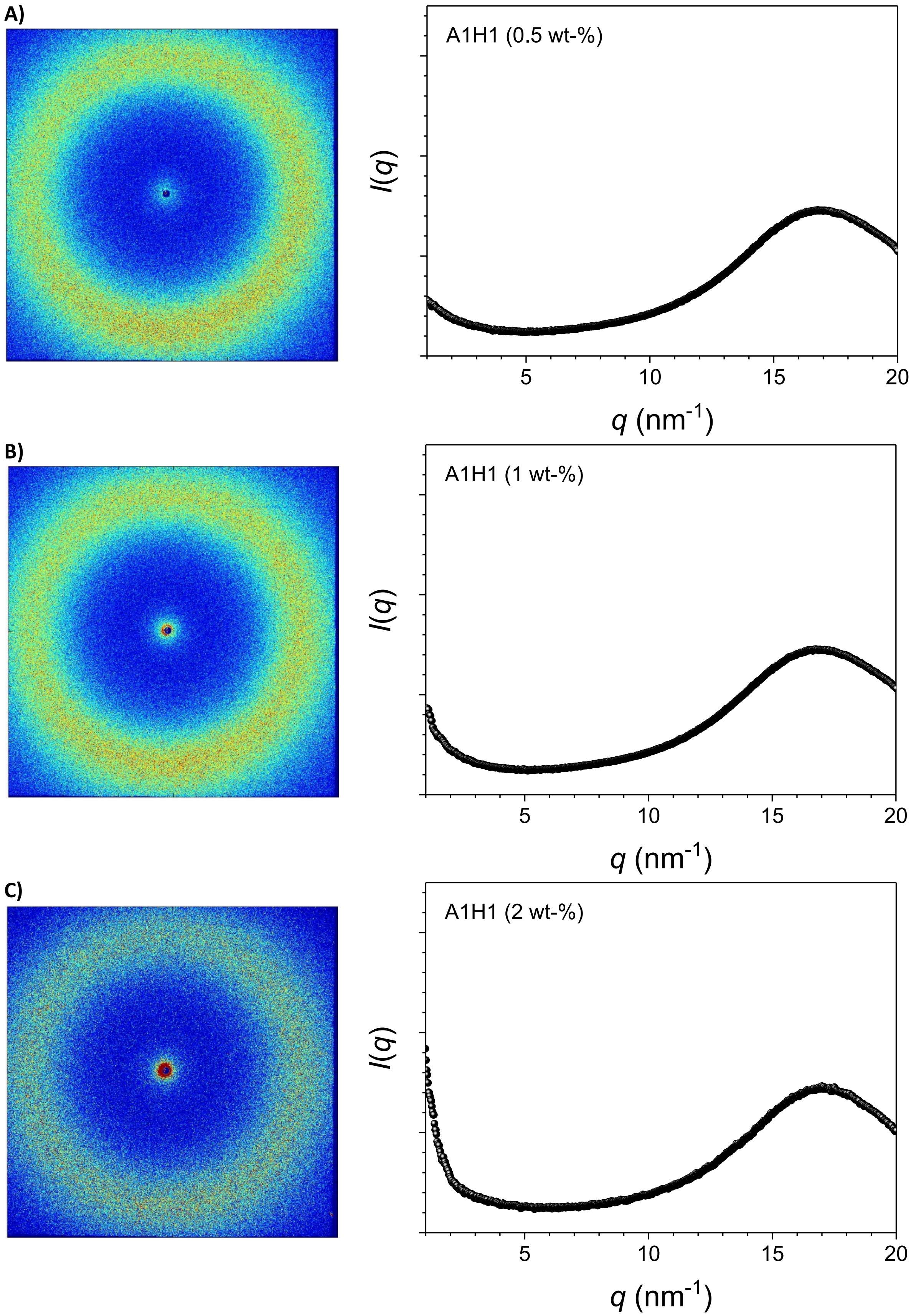


Figure SI-5. 2D and 1D WAXS intensity profile for **A1H1** dispersions in acetonitrile/water at **(A)** 0.5 wt-%, **(B)** 1 wt-%, and **(C)** 2 wt-%.

A) B)

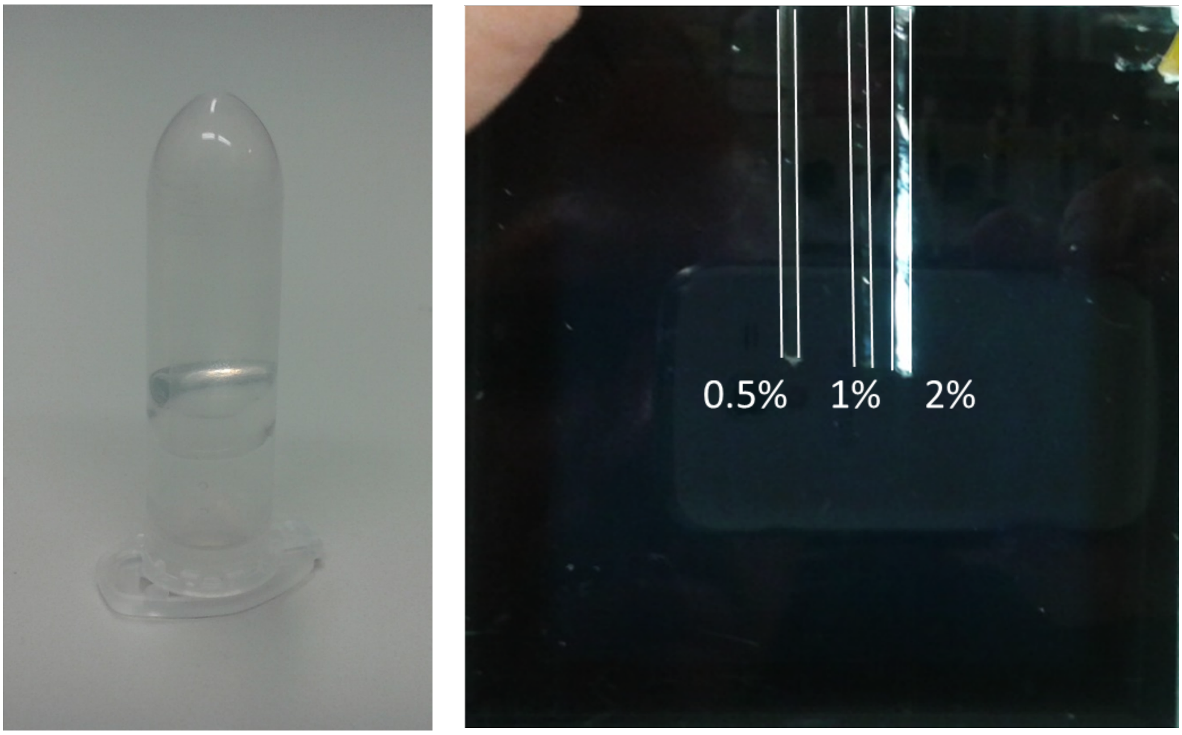


Figure SI-6. (A) Test-tube-inversion method for the 2 wt-% **A1H1** dispersion in acetonitrile/water showing the gel-like characteristic of such sample. **(B)** Polarized light experiment for the 0.5 wt-%, 1 wt-%, and 2 wt-% **A1H1** dispersions in acetonitrile/water. *Note:* the edged of the capillaries are highlighted for a better visualization.

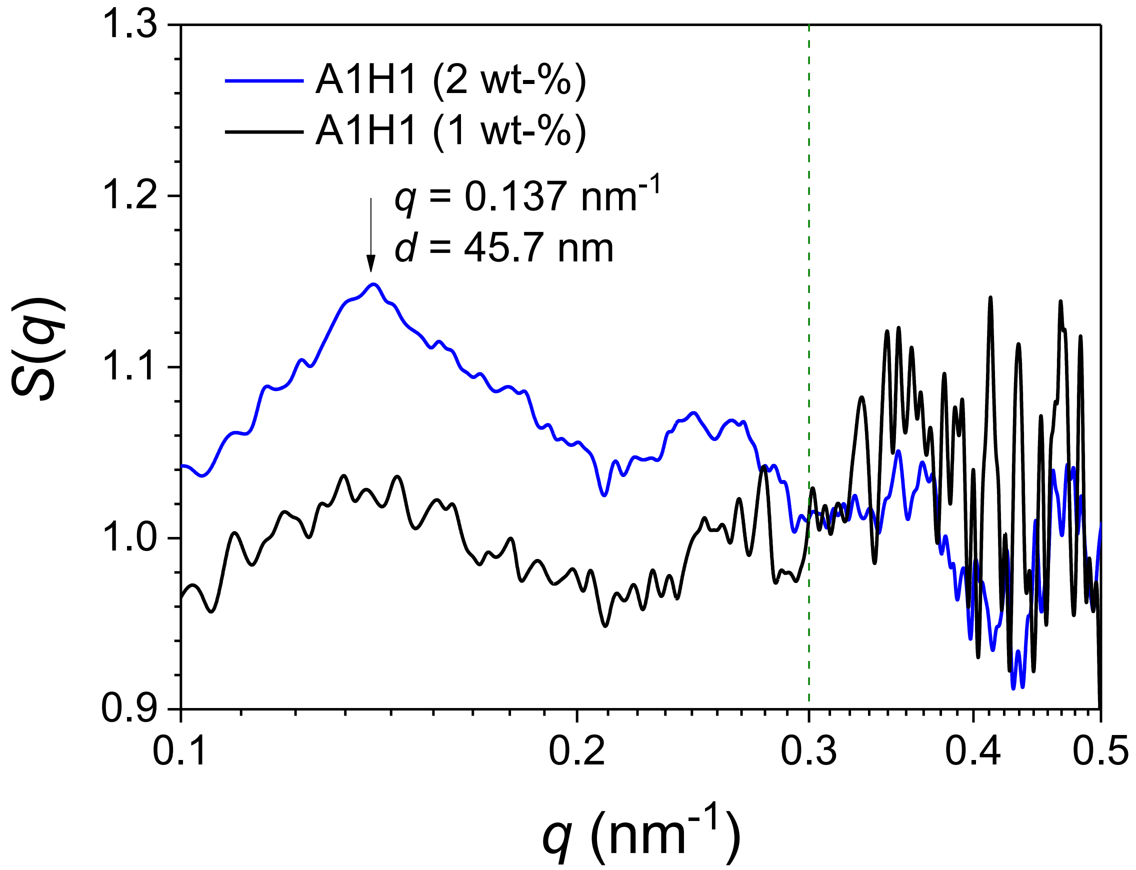


Figure SI-7. Structure factor *S*(*q*) for the non-birefringent (1 wt-%, black curve) and the birefringent (2 wt-%, blue curve) **A1H1** sample in acetonitrile/water dispersion.

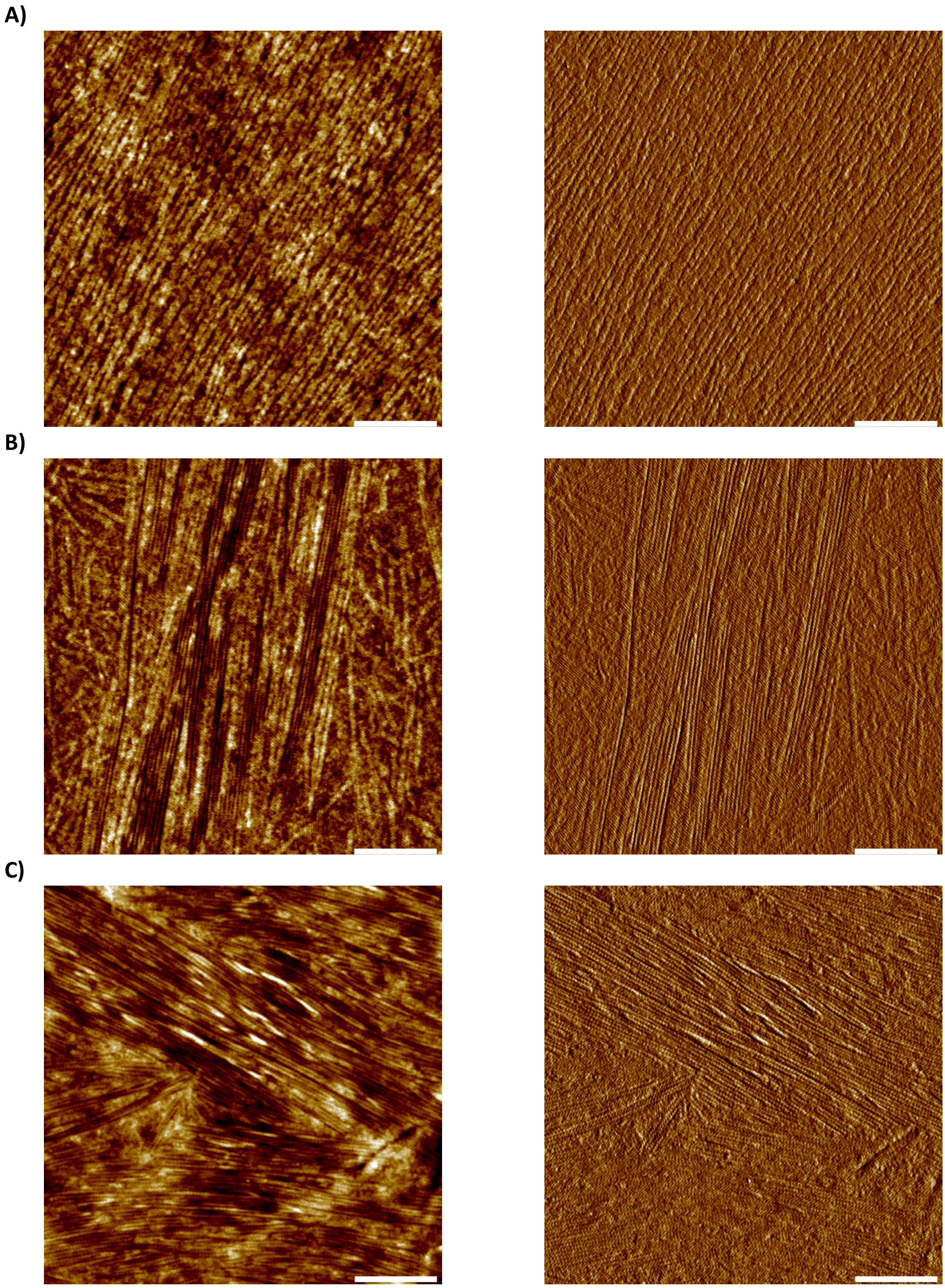


Figure SI-8. AFM height (left) and amplitude (right) profile image of the deposited **(A)** 1 wt-%, **(B)** 2 wt-%, and **(C)** 4 wt-% **A1H1** dispersion in acetonitrile/water. *Note*: scale bar is 300 nm.

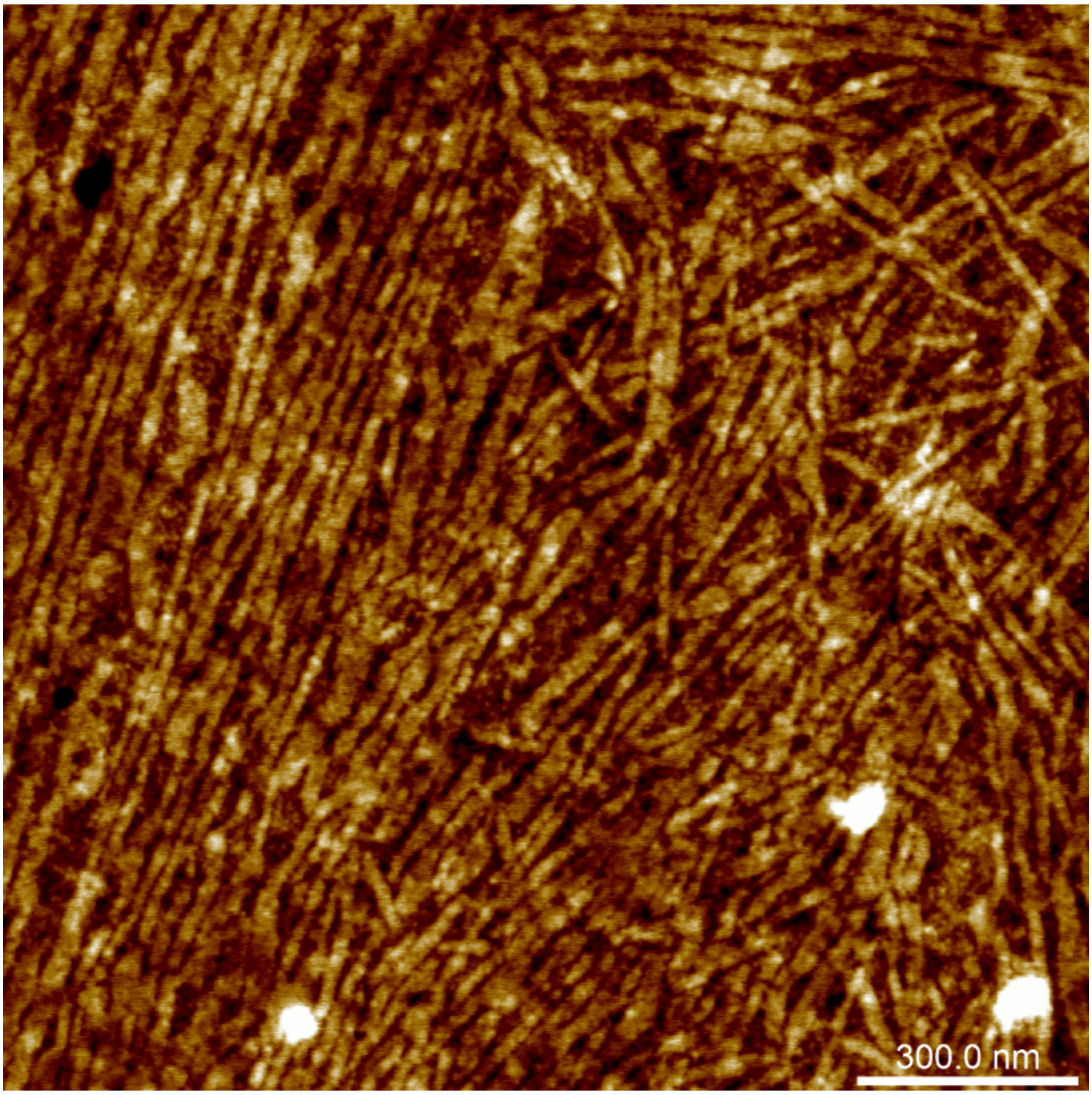


Figure SI-9. AFM height profile image of the deposited 1 wt-% **A1H1** dispersion in acetonitrile/water showing the local alignment due to deposition.

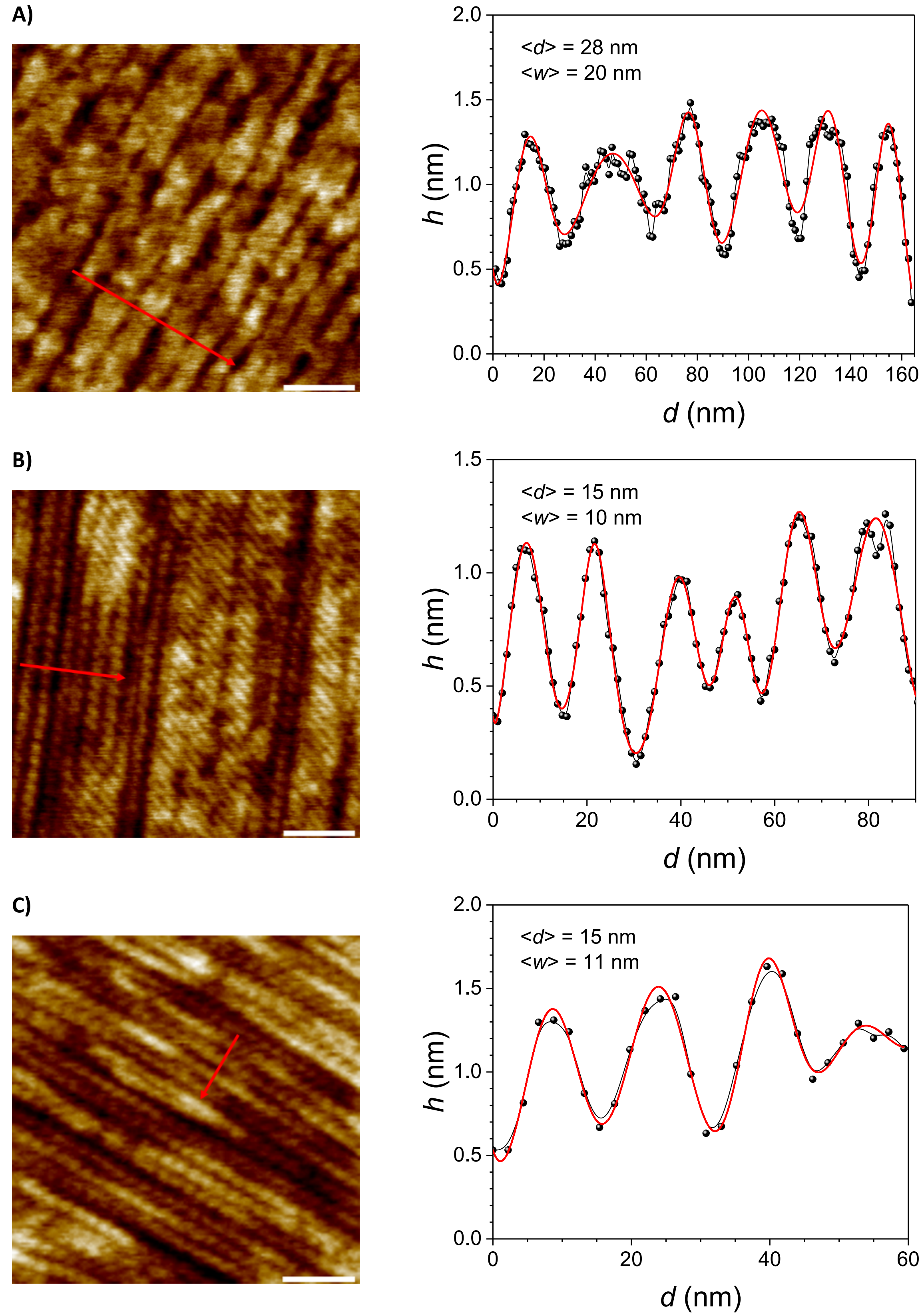


Figure SI-10. AFM height profile image (left) and cross section height profile (right) of the deposited **(A)** 1 wt-%, **(B)** 2 wt-%, and **(C)** 4 wt-% **A1H1** dispersion in acetonitrile/water. *Note*: scale bar is 60 nm.

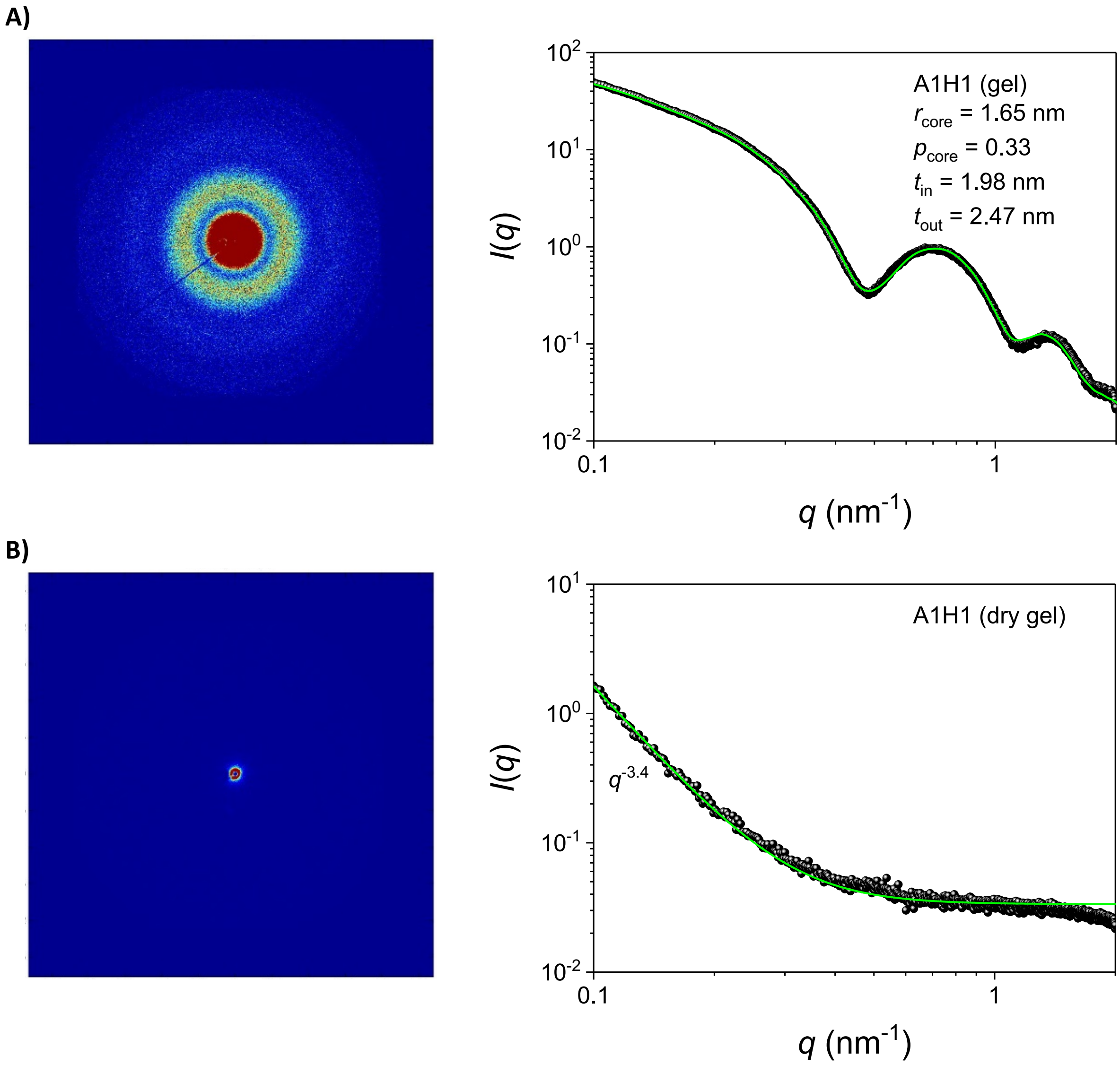
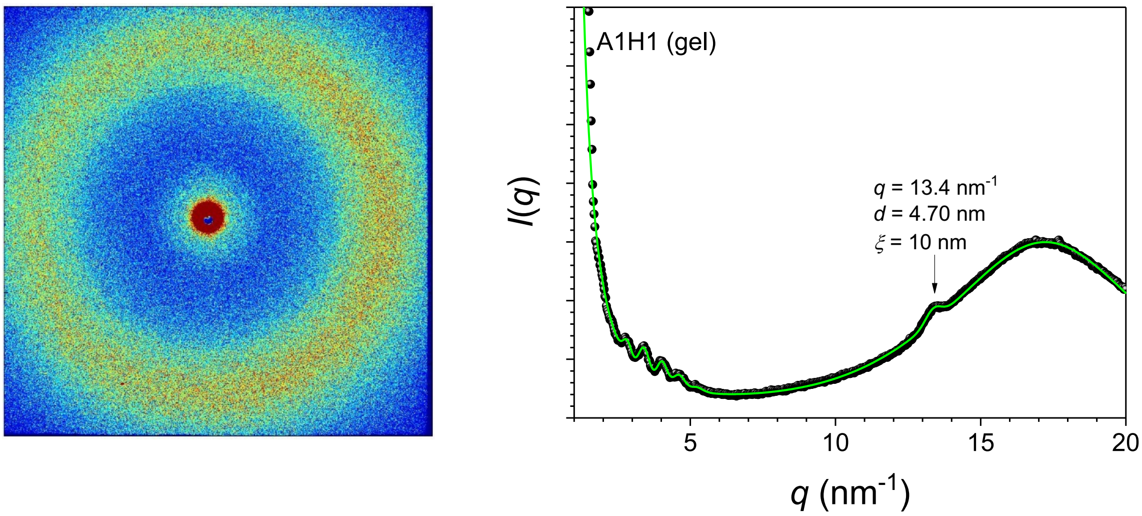


Figure SI-11. 2D and 1D SAXS intensity profiles for **(A)** the **A1H1** gel in acetonitrile/water from the 2 wt-% **A1H1** dispersion, and **(B)** the corresponding dry gel. *Note:* the green fitting curve correspond to the form factor $P(q)$.

A)



B)

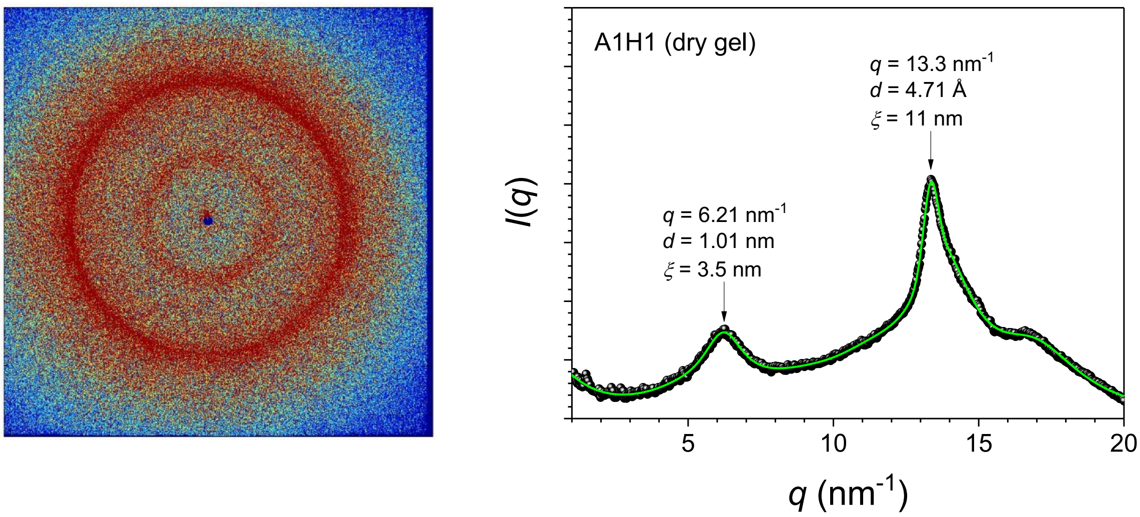


Figure SI-12. 2D and 1D WAXS intensity profiles for **(A)** the **A1H1** gel in acetonitrile/water from the 2 wt-% **A1H1** dispersion, and **(B)** the corresponding dry gel.

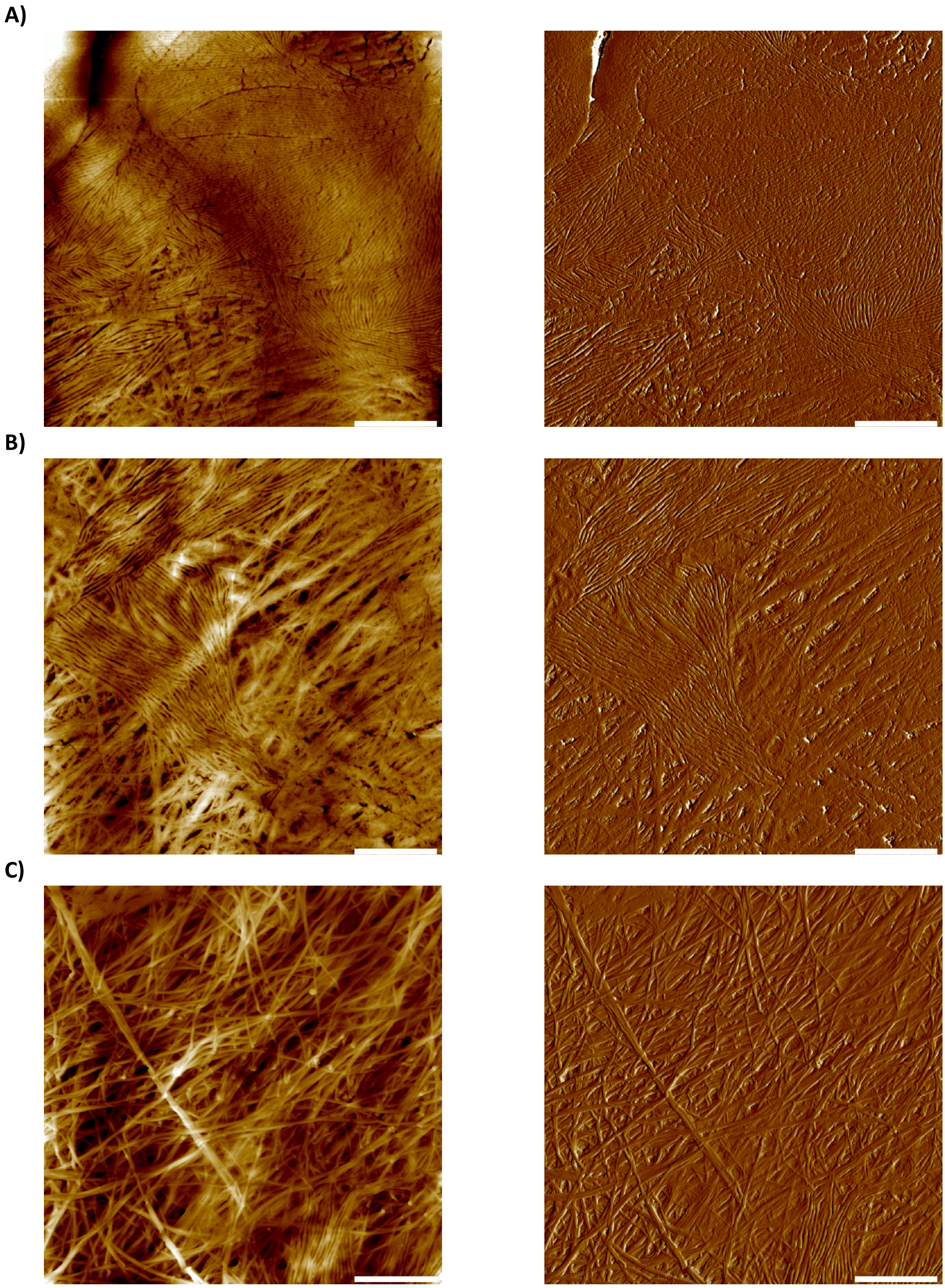


Figure SI-13. AFM height (left) and amplitude (right) profile images of dry gel showing the different type of fibers upon solvent removal. *Note*: scale bar is 1 μm.

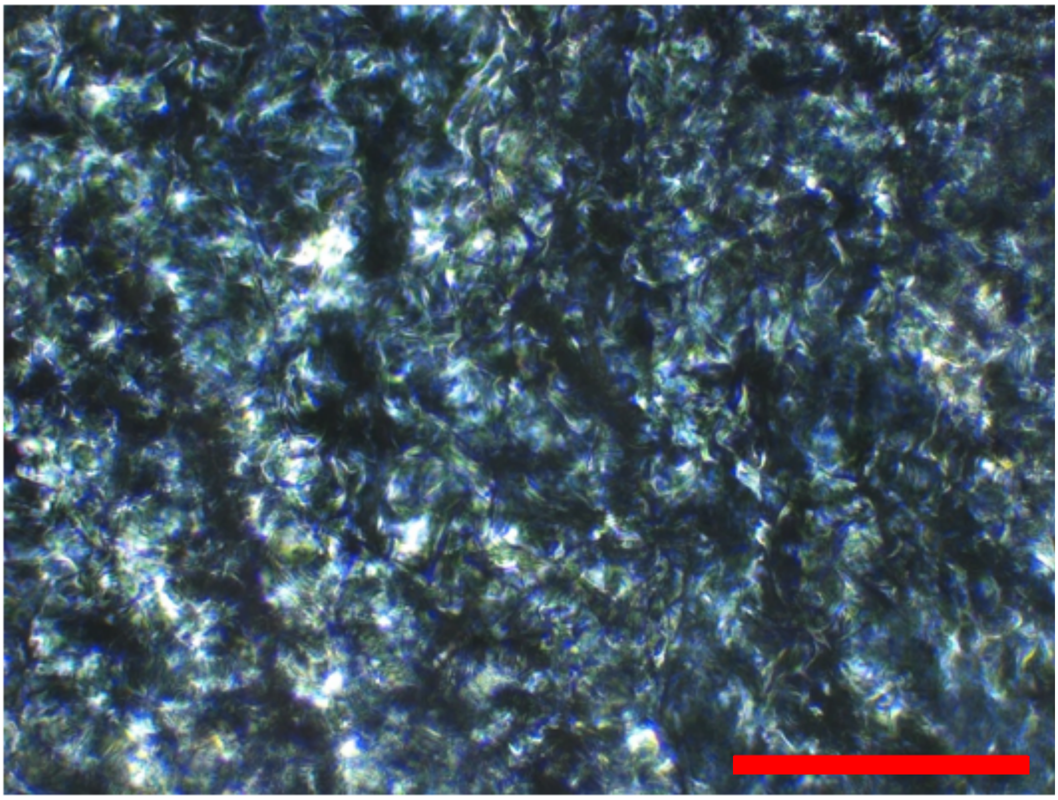


Figure SI-14. Polarized light optical microscope image of the dry gel. *Note*: scale bar is 500 μm

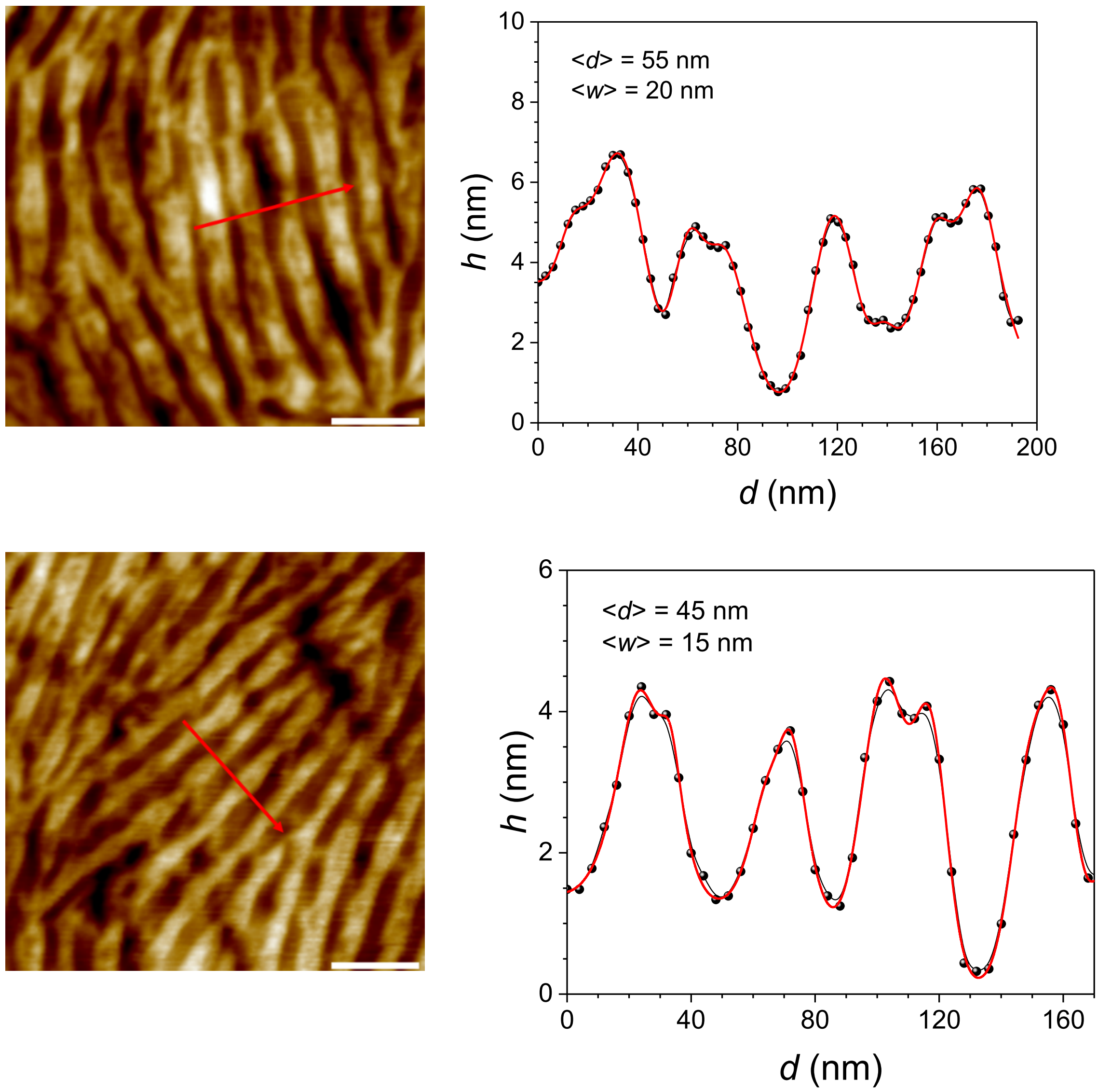


Figure SI-15. AFM height profile images (left) and cross section height profiles (right) of the dry gel. *Note*: scale bar is 100 nm.

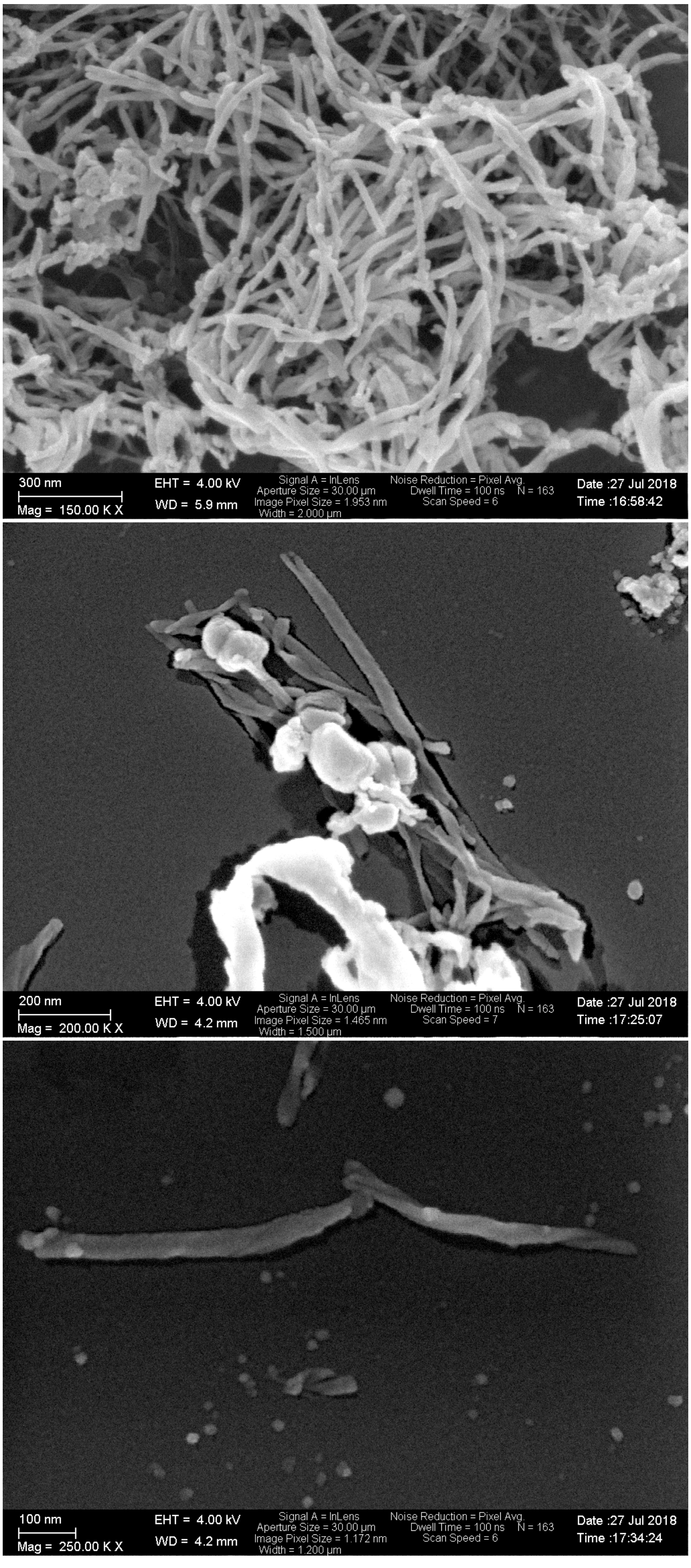


Figure SI-16. CryoSEM images of the deposited 2 wt-% **A1H1** dispersion in acetonitrile/water.

Form factor for a hollow poly-core two-shell cylinder object

The scattering intensity for a colloidal system can be described as $I(q) = NP(q)S(q)$, where *N* is proportional to the concentration and scattering volume, *P*(*q*) is the form factor, and *S*(*q*) is the structure factor.

The form factor for a hollow poly-core two-shell cylinder is described by the following expression:

$$P(q) = k \int_0^{\pi/2} \left[(\rho_{core} - \rho_{in}) V_{core} \frac{2J_1(qr_{core} \sin\alpha)}{qr_{core} \sin\alpha} \frac{sin\left(q\frac{L}{2}cos\alpha\right)}{q\frac{L}{2}cos\alpha} + (\rho_{in} - \rho_{out}) V_{in} \frac{2J_1(qr_{in} \sin\alpha)}{qr_{in} \sin\alpha} \frac{sin\left(q\frac{L}{2}cos\alpha\right)}{q\frac{L}{2}cos\alpha} + (\rho_{out} - \rho_0) V_{out} \frac{2J_1(qr_{out} \sin\alpha)}{qr_{out} \sin\alpha} \frac{sin\left(q\frac{L}{2}cos\alpha\right)}{q\frac{L}{2}cos\alpha} \right]^2 sin\alpha \, \mathrm{d}\alpha$$

Where *k* is the scaling factor, ρ_{core}, ρ_{in}, ρ_{out} and ρ_0 is the scattering length density for the core, inner shell, outer shell and solvent, respectively; V_{core}, V_{in} and V_{out} is the volume of the core, inner shell, and outer shell, respectively; r_{core}, r_{in} and r_{out} is the radius of the core, inner shell and outer shell, respectively; t_{in} and t_{out} is the thickness of the inner shell and outer shell, respectively; and *L* the length of the cylindrical object.

The relationship between the radii and thicknesses are:

$$r_{in} = r_{core} + t_{in}$$

$$r_{out} = r_{core} + t_{in} + t_{out}$$

The corresponding volumes are:

$$V_{core} = \pi r_{core}^2 L$$

$$V_{in} = \pi (r_{core} + t_{in})^2 L$$

$$V_{out} = \pi (r_{core} + t_{in} + t_{out})^2 L$$