

## Supporting information

### Wet spinning and radial self-assembly of a carbohydrate low molecular weight gelator into well organized hydrogel filaments

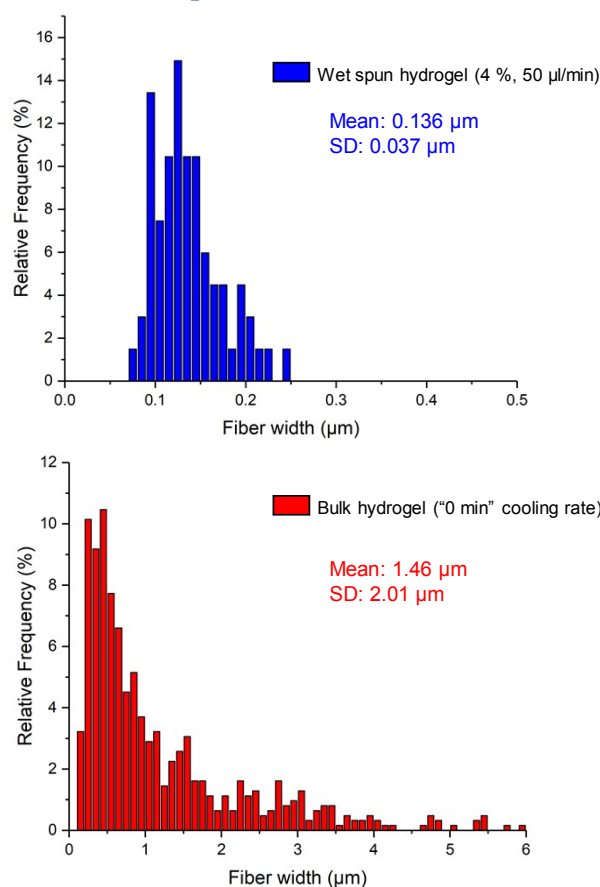
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#### SI-1 Velocity of the jet

| Flow rate                     | 5 $\mu\text{l}/\text{min}$ |               |               | 10 $\mu\text{l}/\text{min}$ |                |                |
|-------------------------------|----------------------------|---------------|---------------|-----------------------------|----------------|----------------|
| Replicate n°                  | 1                          | 2             | 3             | 1                           | 2              | 3              |
| Measured jet diameter (mm)    | 0.168                      | 0.176         | 0.185         | 0.146                       | 0.184          | 0.222          |
| Measured jet velocity (mm/s)  | $8.6 \pm 0.5$              | $8.3 \pm 0.5$ | $8.1 \pm 0.4$ | $10.5 \pm 0.4$              | $10.4 \pm 0.5$ | $11.1 \pm 0.8$ |
| Predicted jet velocity (mm/s) | $3.8 \pm 0.9$              | $3.4 \pm 0.8$ | $3.1 \pm 0.7$ | $10 \pm 3$                  | $6 \pm 1$      | $4.3 \pm 0.8$  |

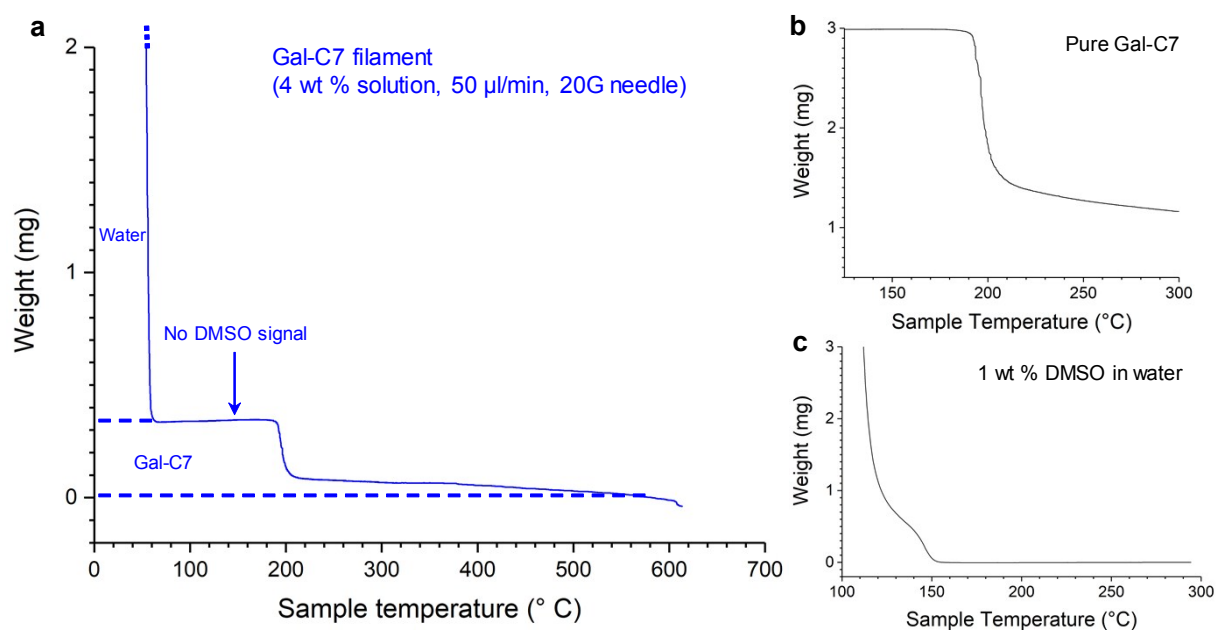
**Table SI-1:** velocity of the jet in different conditions

#### SI-2- Fiber width distribution: comparison between bulk and wet spun hydrogels



**Figure SI-2:** Fiber width distribution: comparison between wet spun (top) and bulk (bottom) hydrogels

### SI-3- Thermogravimetric analysis (TGA)



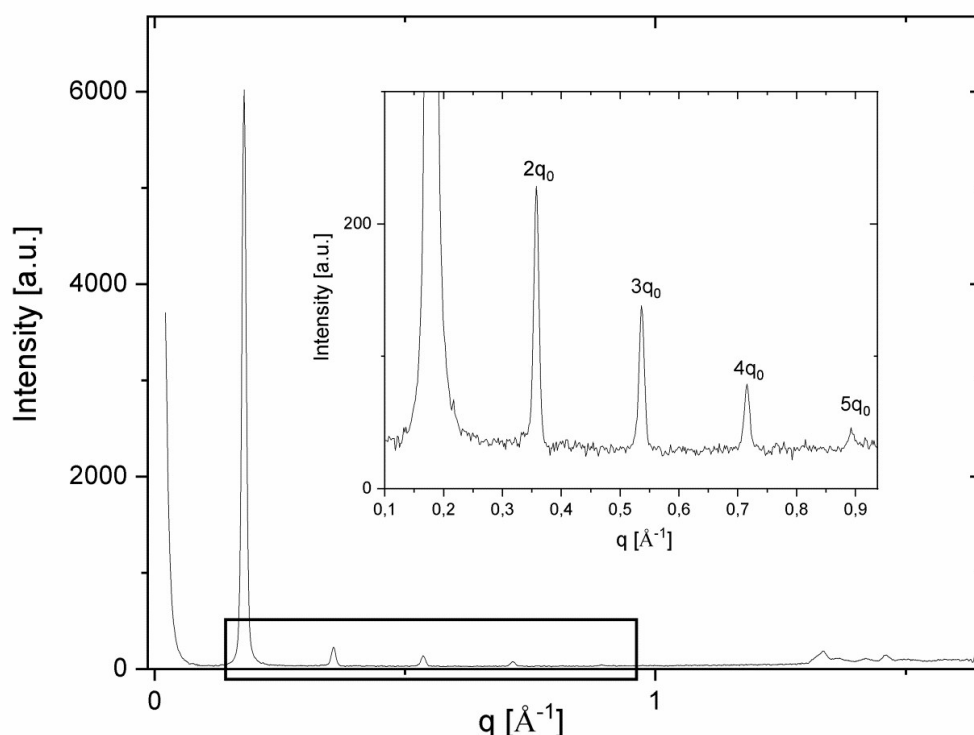
**Figure SI-3:** (a) Magnification on the thermogram of a wet-spun Gal-C7 filament. The arrow indicates the temperature expected for the DMSO transition. (b) Magnification of the first transition of pure Gal-C7. (c) Magnification of the thermogram of a solution of water containing 1 wt % of DMSO.

### SI-4- Lifetime of the filaments

| Flow rate    | 25 µl/min                      |                                  | 50 µl/min                      |                                  |
|--------------|--------------------------------|----------------------------------|--------------------------------|----------------------------------|
| Conditions   | Lifetime exposed to air (days) | Lifetime under cover slip (days) | Lifetime exposed to air (days) | Lifetime under cover slip (days) |
| Water (RT)   | 3                              | 5                                | 2                              | 3                                |
| DMEM (RT)    | > 31                           | > 31                             | > 34                           | > 34                             |
| DMEM (37 °C) | 7                              | 4                                | < 1                            | 4                                |
| PBS (RT)     | < 1                            | 3                                | < 1                            | 2                                |

**Table SI-4:** Lifetime of the filaments in different conditions

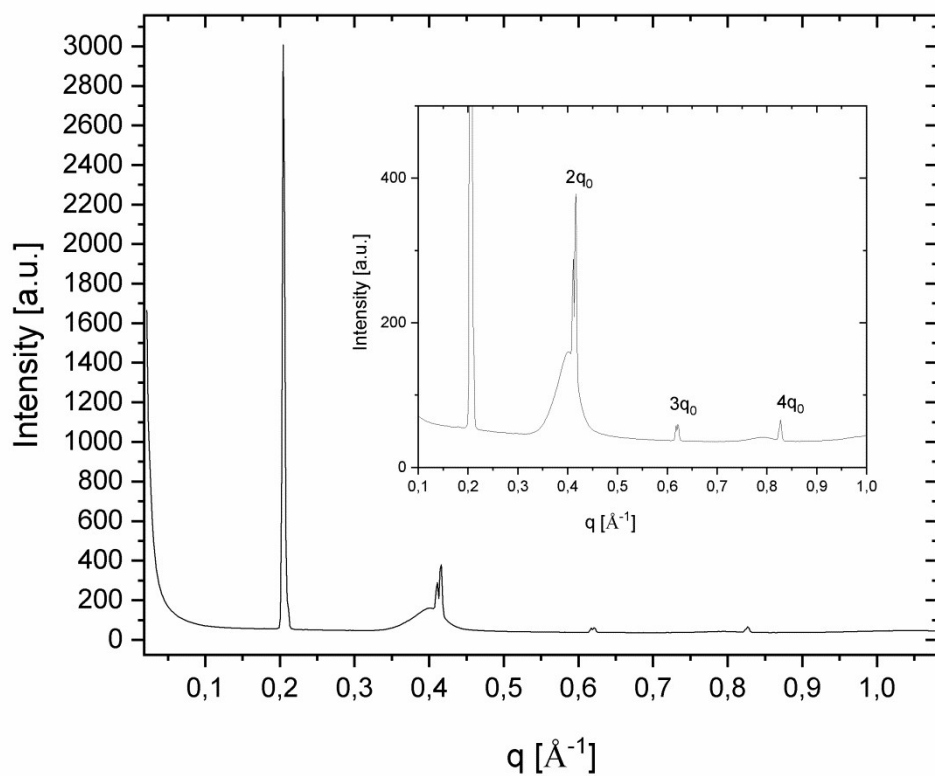
## SI-5- Small Angle X-Ray Scattering (SAXS)



**Figure SI-5a.** SAXS intensity as a function of the scattering vector of the fresh wet-spun Gal-C7 gel

The SAXS curve for the wet-spun gel is reported in Figure SI-5a. The main feature is the presence of a strong scattering peak followed by four less intense ones at positions  $2q_0$ ,  $3q_0$ ,  $4q_0$  and  $5q_0$ , indicative of a well defined lamellar organization. From the position of the first order peak ( $q_0^* = 0.179 \text{ Å}^{-1}$ ) we can calculate the long period spacing of the lamellar structure,  $d = 35.1 \text{ Å}$ . The fiber width estimated from SAXS spectrum is 60 nm from the peak width (ref). Besides, the low  $q$  region of the curve presents a power law trend,  $I(q) \propto q^{-4}$ , which corresponds to the Porod region for the sharp interface of the fibers observed in cryo-SEM images.

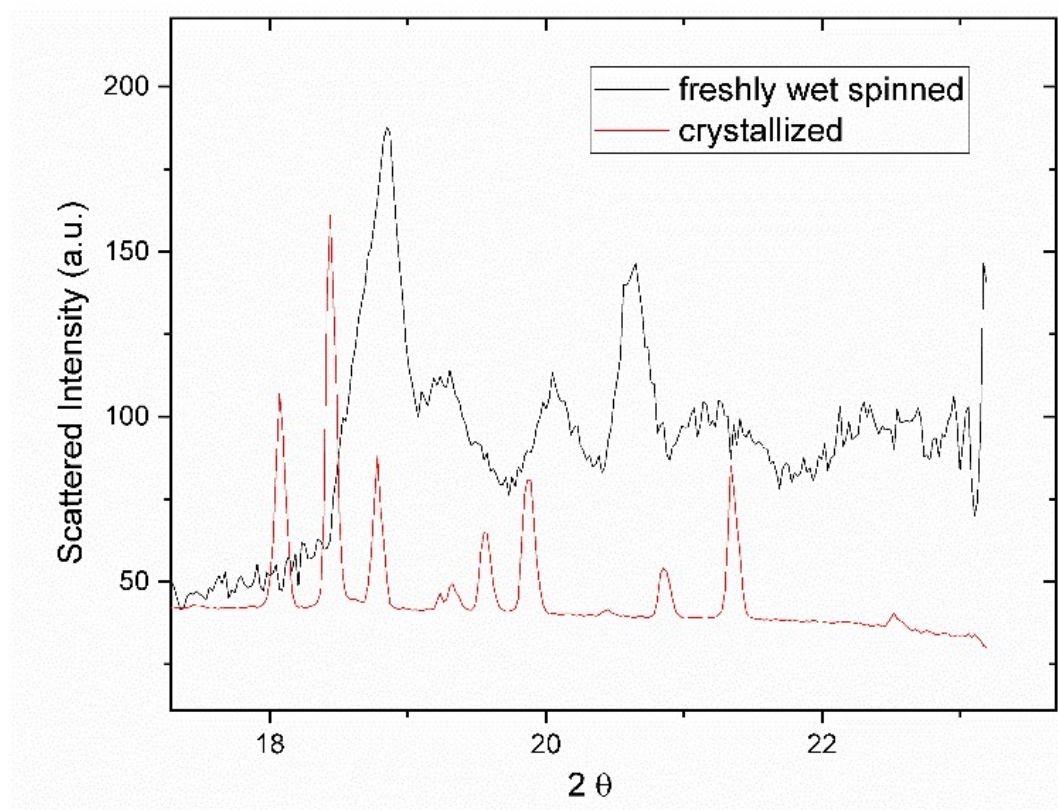
| Sample                                              | $q_0 [\text{Å}^{-1}]$ | $[\text{Å}]$  |
|-----------------------------------------------------|-----------------------|---------------|
| Fresh wet-spun gel threads                          | 0.179                 | 35.1          |
| Crystals                                            | 0.204                 | 30.5          |
| Gel prepared by slow cooling of an aqueous solution | 0.178 and 0.164       | 35.2 and 38.3 |



**Figure SI-5b.** SAXS intensity as a function of the scattering vector of the Gal-C7 crystals.

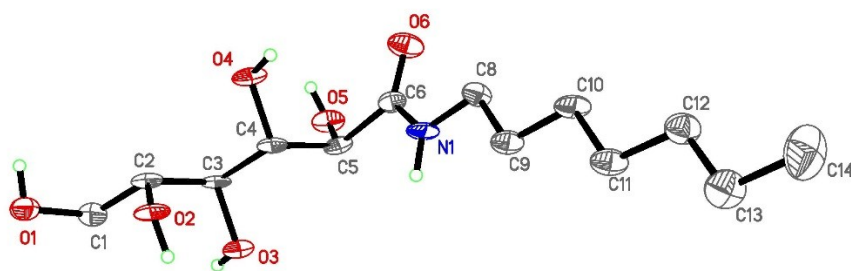
The obtained crystals were recovered and analysed by SAXS and the corresponding spectrum is reported in Figure SI-5b. The SAXS region looks very similar to that of the wet-spun filament, a lamellar order is recognized with a 30.5  $\text{\AA}$  spacing (peak position, 0.204  $\text{\AA}^{-1}$ ).

## SI-6- Wide Angle X-Ray Scattering (WAXS)



**Figure SI-6.** WAXS intensity as a function of the scattering vector of the crystallized and freshly wet-spun Gal-C7.

## SI-7- X-Ray diffraction



**Figure SI-7a :** Molecular view of Gal-C7 in the crystal. Thermal ellipsoids represent 50% probability level. H atoms (except for that on O and N atoms) are omitted for clarity.

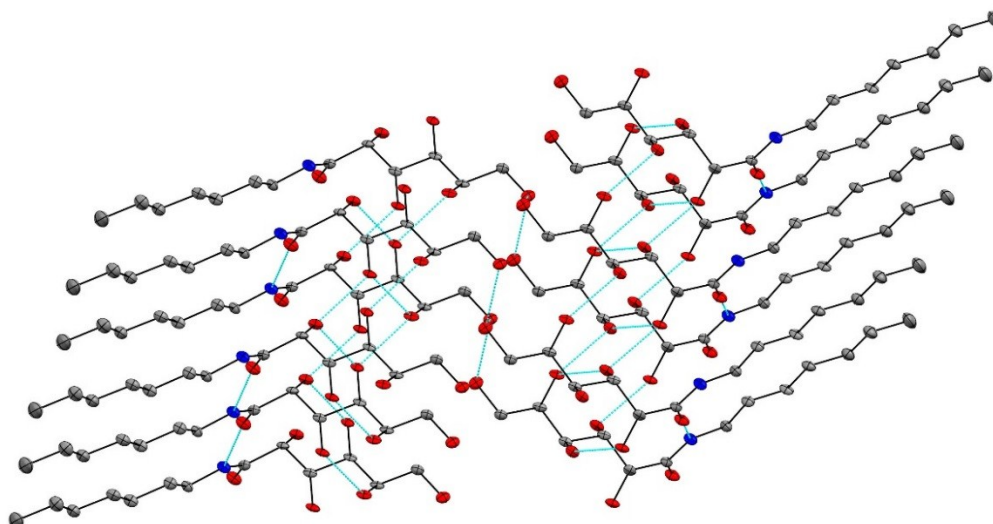
| D-H...A             | d(D-H)  | d(H...A) | d(D...A) | <(DHA) |
|---------------------|---------|----------|----------|--------|
| O(1)-H(1)...O(1)#1  | 0.85(8) | 2.05(8)  | 2.894(5) | 170(8) |
| O(2)-H(2)...O(4)#2  | 0.89(9) | 2.00(8)  | 2.738(7) | 139(7) |
| O(3)-H(3)...O(2)#3  | 0.83(9) | 1.90(9)  | 2.722(7) | 167(9) |
| O(4)-H(4)...O(5)#4  | 0.86(8) | 1.85(8)  | 2.700(7) | 171(8) |
| O(5)-H(5)...O(3)#5  | 0.78(8) | 2.07(8)  | 2.760(7) | 147(8) |
| N(1)-H(1N)...O(6)#2 | 0.89(7) | 2.06(7)  | 2.943(7) | 172(6) |

Symmetry transformations used to generate equivalent atoms:

#1  $-x+2, y-1/2, -z+1$  #2  $x, y+1, z$  #3  $x+1, y, z$

#4  $x-1, y, z$  #5  $x, y-1, z$

**Table SI-7.** Hydrogen bonds for Gal-C7 in the crystal [ $\text{\AA}$  and  $^\circ$ ].



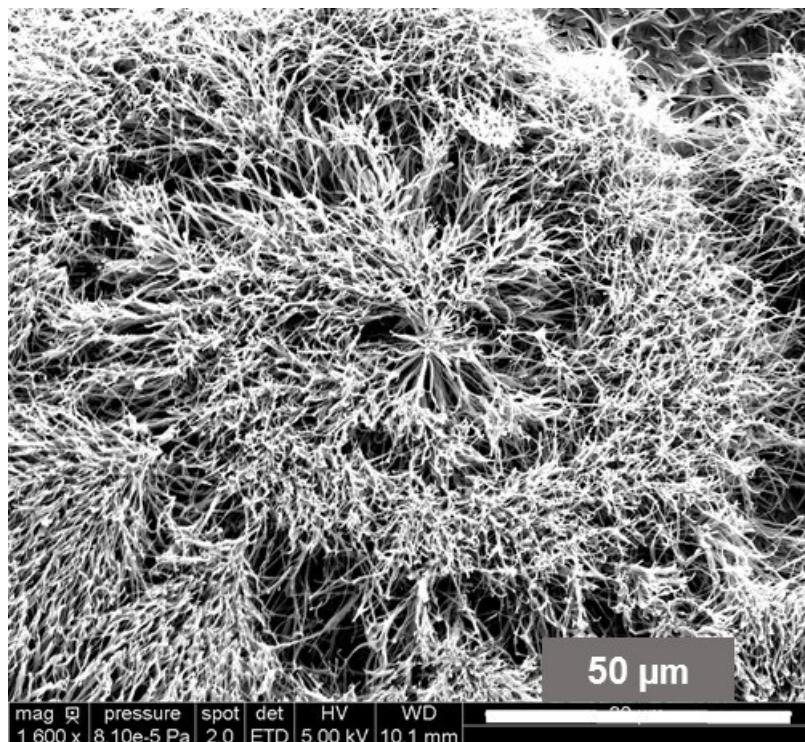
**Figure SI-7b:** Representative O-H---O and N-H---O bonds are drawn as dotted lines (H atoms are omitted for clarity).



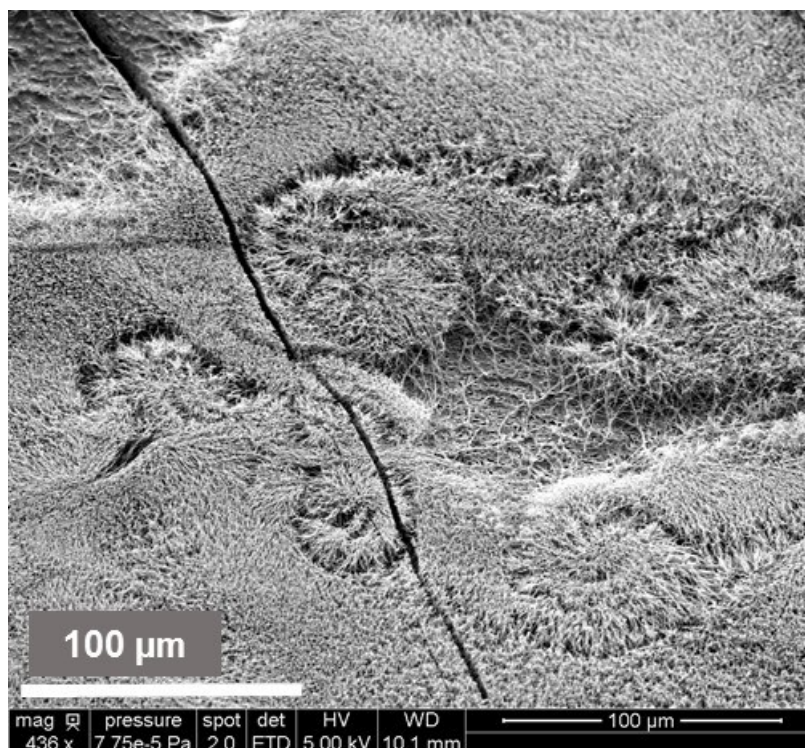
## SI-8- Cryo-Scanning Electronic Microscopy – Extra images

*Spirals in filaments produced with a 5wt% solution at 100  $\mu\text{L}/\text{min}$*

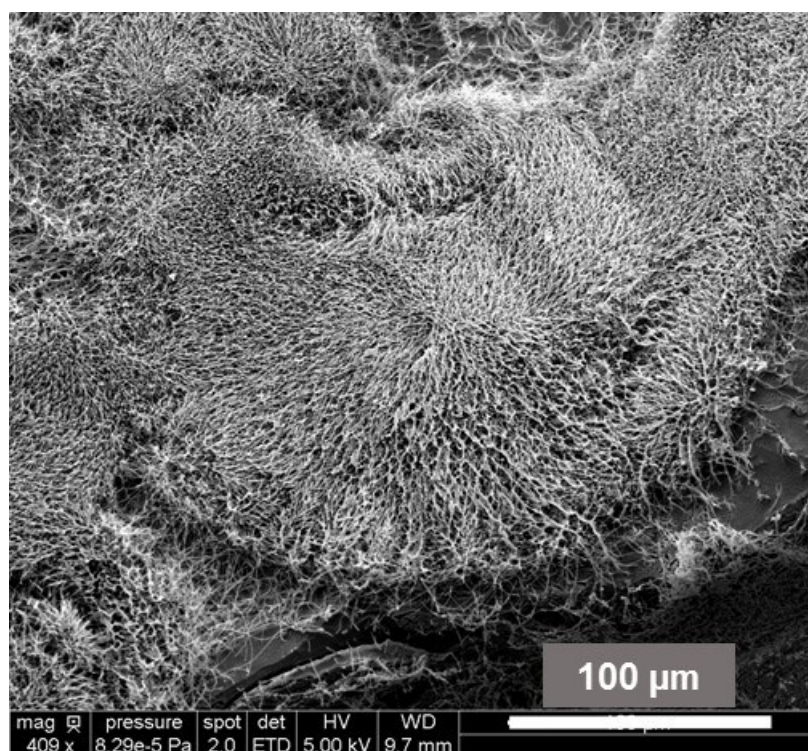
Enlargement of Figure 7e



Other spirals



*Radial organization, 5wt%, 100 $\mu$ L/min*



*Radial organization, 5wt%, 50 $\mu$ L/min*

