Electronic Supplementary Information (ESI)

Polymer-free electrospinning of tannic acid and cross-linking in water for hybrid smart supramolecular nanofibres

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Estimation of hydrodynamic diameters D_{TA} of TA aggregates from DLS measurements:

Hydrodynamic diameters D_{TA} of TA aggregates were calculated from DLS measurements and the following equations:
Scattering vector q:

$$q = \frac{4\pi n}{\lambda}\sin\left(\frac{\theta}{2}\right)$$

With n the refractive index of the solvent, λ = 633 nm is the wavelength of the used laser and θ the used scattering angle.

Diffusion coefficient:

$$D = \frac{1}{2\tau q^2}$$

With τ the characteristic time of the fast mode obtained from DLS measurements. They were obtained from the field correlation functions which were fitted with the stretched exponential functions having one to three relaxation modes. These functions fit well empirically the autocorrelation functions.[1]

The diameter D_{TA} of TA aggregates are estimated from:

$$D_{TA} = \frac{2k_B T}{6\pi\eta D}$$

With k_B = 1,38 × 10^{-23} $m^2.kg.s^{-2}.K^{-1}$ the Boltzmann constant, T = 298 K the temperature and η the viscosity of the solvent.

The following parameters were measured to calculate D_{TA}:

C_{TA} (wt%)	τ in water (ms)	τ in water/ethanol 37.5/62.5 wt% (ms)
2	1.91 x 10^{-2}	3.53 x 10^{-2}
10	4.89 x 10^{-2}	1.30 x 10^{-1}
35	1.0 x 10^{-1}	2.03 x 10^{-1}

Solvent	water	Water/ethanol 37.5/62.5 wt%
n	1.332	1.36
η (mPa.s)	0.89	2.22

[1] Nisato, G.; Hebraud, P.; Munch, J.-P.; Candau, S. *J.´ Phys. Rev. E*, 2000, **061**, 02879.

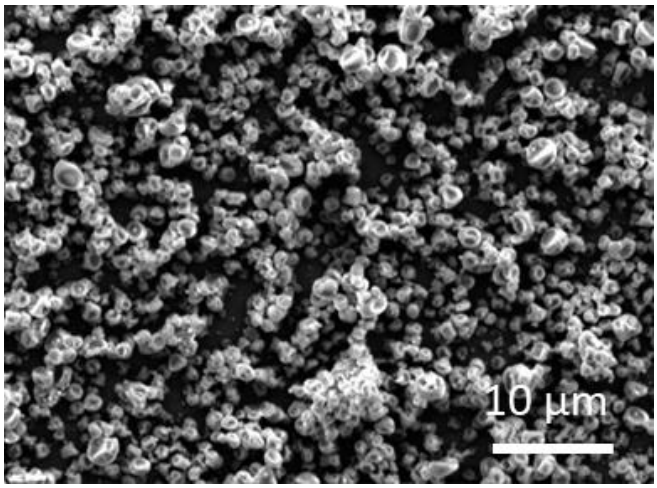


Figure S1: Electrospinning of 100 mg/mL of TA in ethanol.

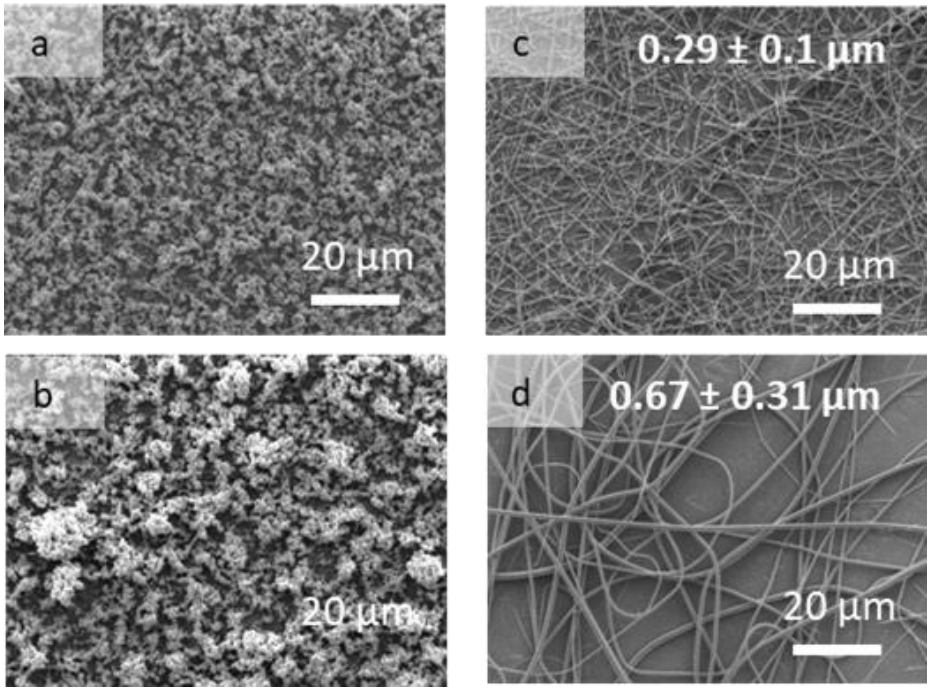


Figure S2: SEM images of TA fibers obtained in water-ethanol mixtures (87.5-12.5 wt%) at concentrations of a) 40 wt%, b) 45 wt%, c) 50wt%, d) 55 wt% and their average fiber diameters.

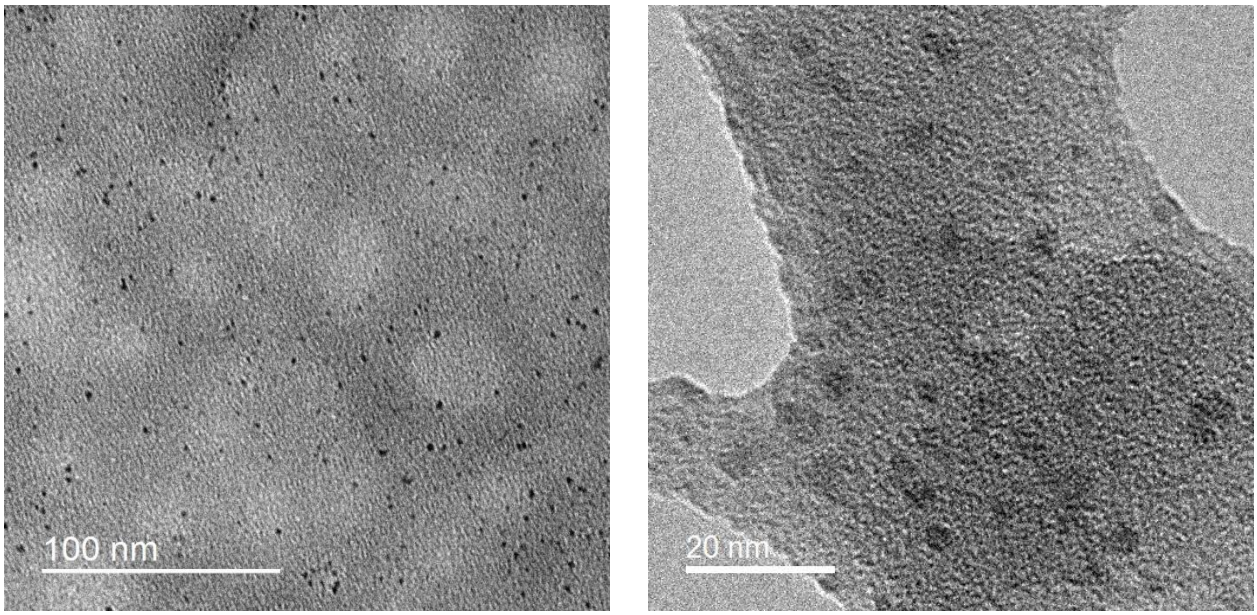


Figure S3: Cryo-TEM picture obtained from a TA solution in water-ethanol at 37.5-62.5 wt% and for CTA = 2 wt% showing dark domains of TA aggregates.

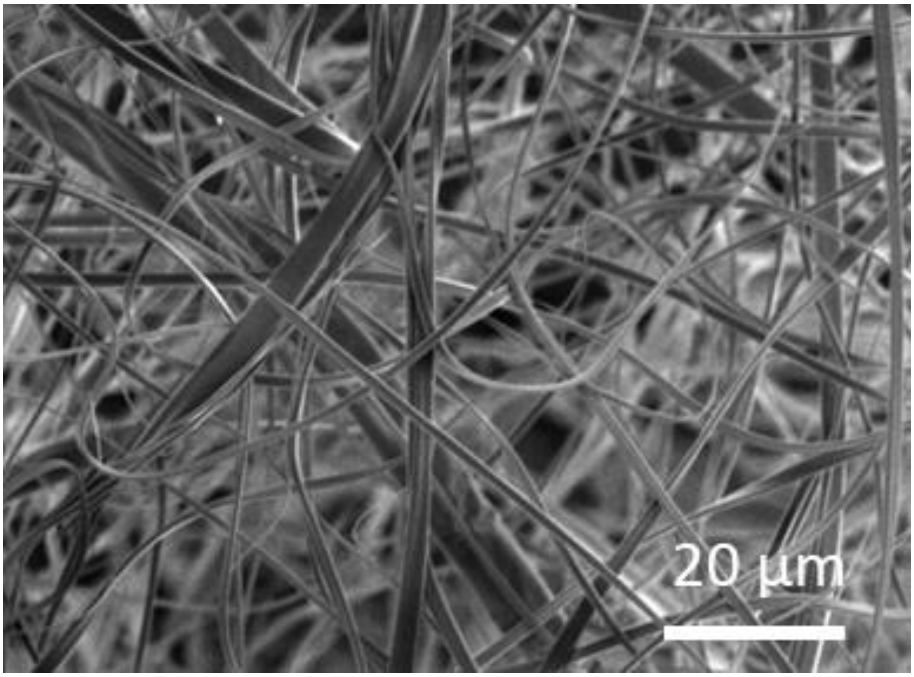


Figure S4: SEM picture of the fibres obtained in the water-ethanol mixture 37.5-62.5 wt% at concentration C_{TA}= 45 wt% which were used before cross-linking.

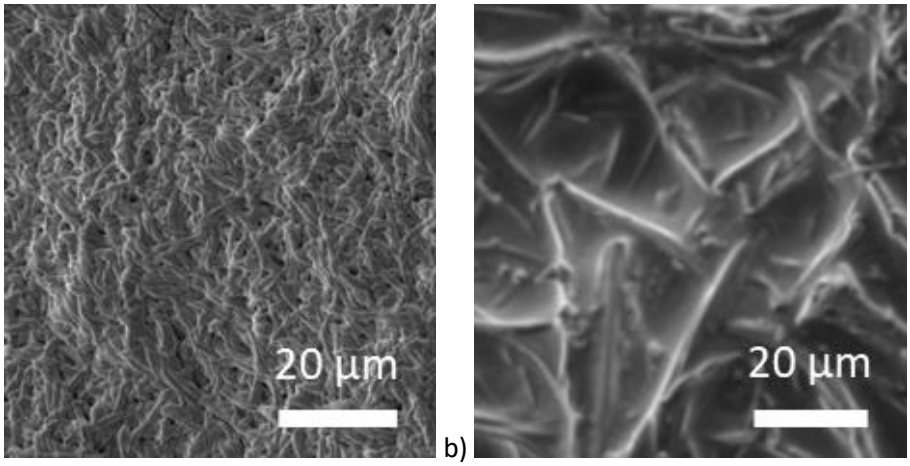


Figure S5: SEM picture of the fibres obtained just after crosslinking in aqueous $NaIO_4$ solution a) and Fe(III) solution b).

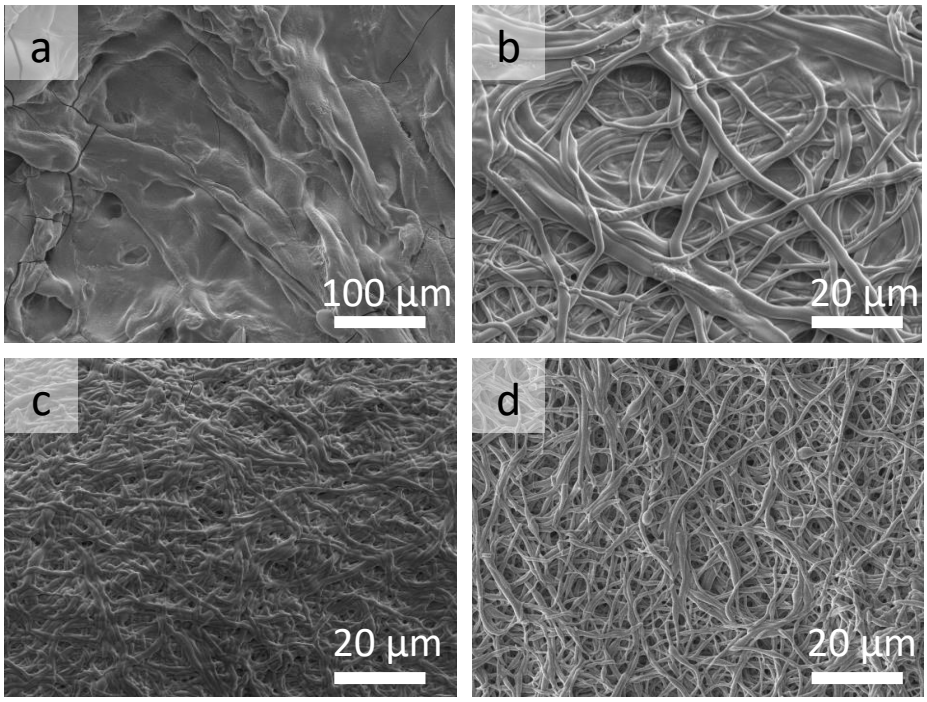


Figure S6: SEM picture of the fibres obtained after crosslinking in aqueous $NaIO_4$ solution of various concentrations for 10 s followed by washing for 6 hours in distilled water. a) 3 wt%, b) 5 wt%, c) 7 wt%, d) 9 wt%.

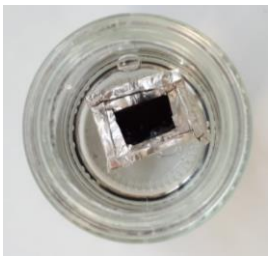
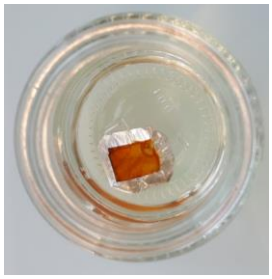
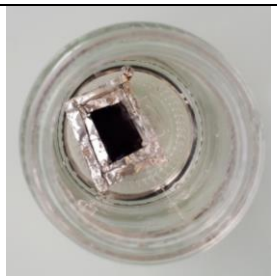
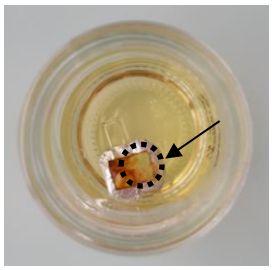
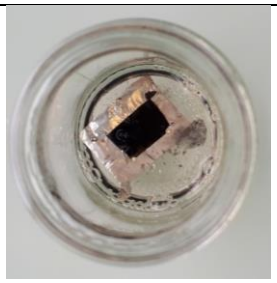
Duration in water	Crosslinking with Fe^{3+}		Crosslinking with $NaIO_4$	
1 day		The scaffold is still intact. The scaffold is still intact. The scaffold is black due to oxidation of the galloyl groups by Fe^{III} into quinones and formation of covalent bounds.		The scaffold is still intact
3 days		The scaffold is still intact		The scaffold is degrade: a hole appeared (arrow)
7 days		The scaffold is still intact	No picture	The scaffold is completely degraded

Figure S7: Pictures showing the evolution of crosslinked scaffolds after several days in water.

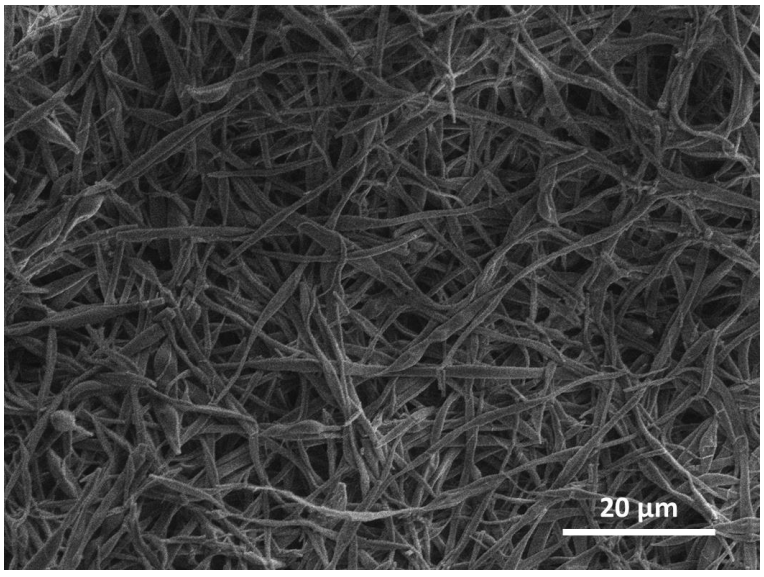


Figure S8: SEM picture of the crosslinked fibres with Fe(III) after 7 days in water.