Electronic Supplementary Material (ESI) for Materials Chemistry Frontiers.

Supporting Information – Materials Chemistry Frontiers

Lightweight foams of amine-rich organosilica and cellulose nanofibrils by foaming and controlled condensation of aminosilane

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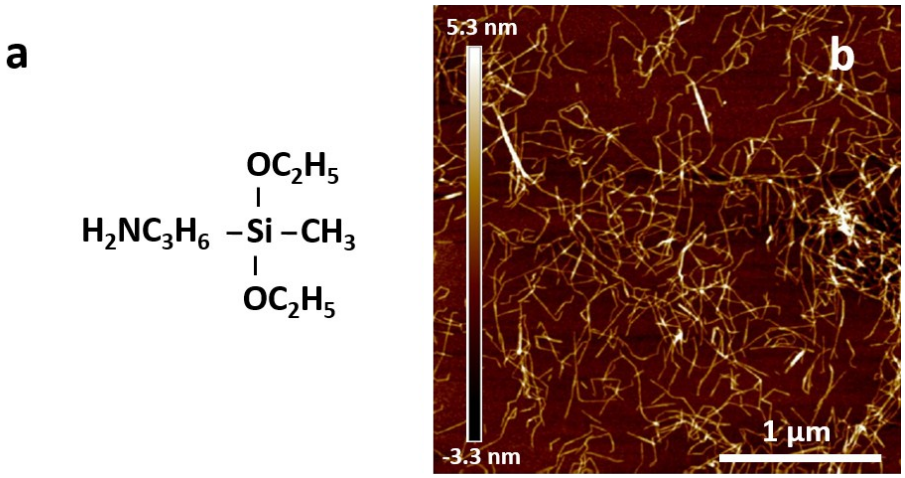
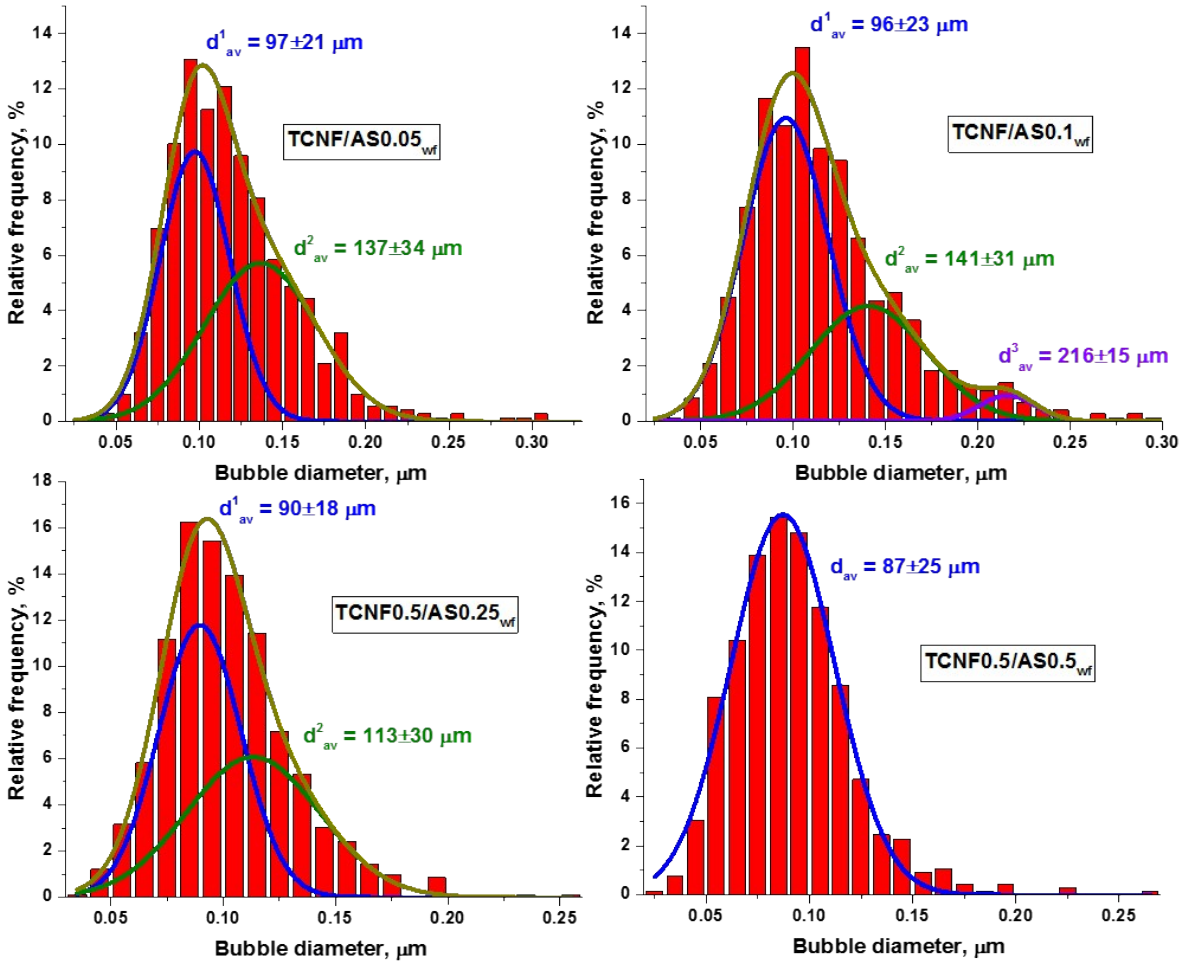
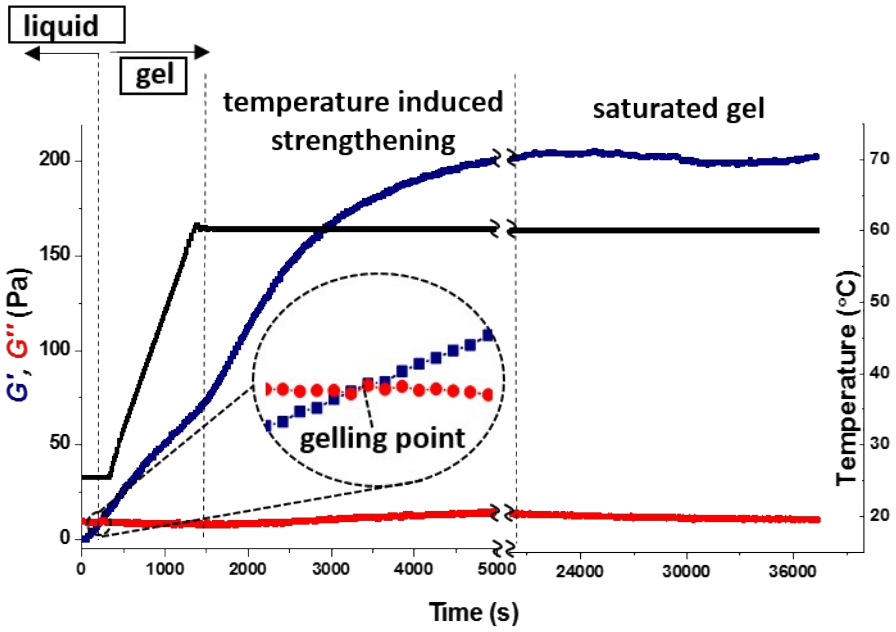


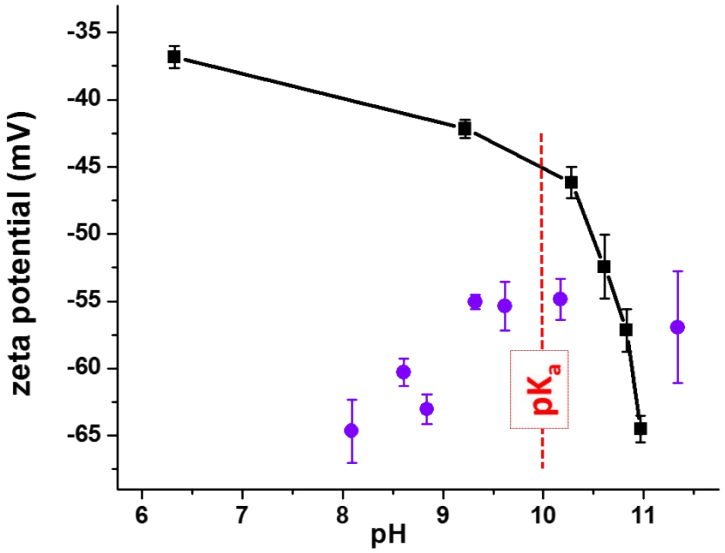
SI.1. Materials for the preparation of solid composite siliceous foams: (a) chemical structure of 3-aminopropyl(methyl)diethoxysilane (AS). (b) Topography image of TCNFs.



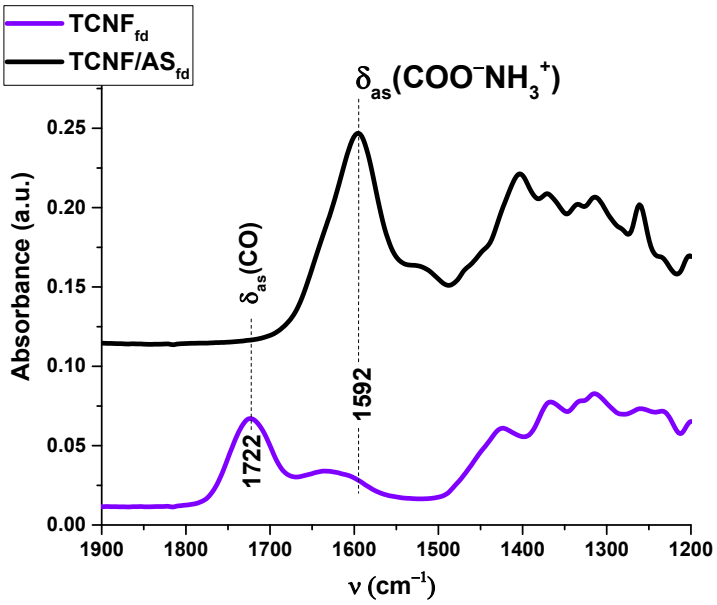
SI.2. Distributions of the air bubble diameters in TCNF/AS_{wf}, measured by optical microcamera right after foaming: *upper left* TCNF/AS0.05, *upper right* TCNF/AS0.1, *bottom left* TCNF/AS0.25 and *bottom right* TCNF/AS0.5. The distributions were fitted with a Gaussian function and the average diameters are presented for the corresponding Gaussian peak on each histogram.



SI.3. Evolution of the storage (*G'*, blue curve) and loss (*G''*, red curve) moduli over time for TCNF/AS3$_{wf}$. The wet foam was heated at 60˚C (heating rate of 2˚C/min) and kept at this constant temperature overnight. The inset zooms in on the gelling point.



SI.4. Zeta potential of a dilute aqueous dispersion of TCNFs (purple dots) and of the TCNF/AS1.5$_d$ (black squares) as a function of the pH. The red dashed line represents the pK$_a$ (=10) value of neat AS.



SI.5. ATR FTIR spectra of $TCNF_{fd}$(pH 4.5) with carboxylic acid groups (–COOH) (bottom purple curve) and $TCNF/AS_{fd}$(pH 9.6) (top black curve) with equimolar content of AS amino-groups and TCNFs carboxylic groups. The TCNFs were ion exchanged from Na^+ to H^+ using a 0.5 M HCl solution.

Table SI.1. Relative integral area for the bands of AS monomers, dimers and polymers determined from the liquid state {1H}^{29}Si NMR of AS and TCNF/AS dispersions at different temperature, pH, and time.

Dispersions and Foams	Integral area for AS monomer	Integral area for AS dimer or polymer end-groups	Integral area for organosilica polymer
AS3d at 20 ˚C after 4 h	1	0.42	0.05
AS3d at 20 ˚C after 16 h	1	0.43	0.03
TCNF/AS3d(11.1) at 20 ˚C after 4 h	1	0.42	0.03
TCNF/AS3d(11.1) at 20 ˚C after 16 h	1	0.43	0.03
TCNF/AS3d(11.1) at 60 ˚C after 4 h	1	0.43	0.02
TCNF/AS3d(11.1) at 60 ˚C after 16 h	1	0.48	0.03
TCNF/AS3d(11.1) at 60 ˚C with in situ evaporation after 4 h	1	0.52	0.03
TCNF/AS3d(11.1) at 60 ˚C with in situ evaporation after 16 h	1	0.51	0.08
TCNF/AS3d(11.1) at 60 ˚C with pre-evaporation (60 ˚C, overnight) after 4 h	1	1.49	0.65
TCNF/AS3d(10.7) at 60 ˚C after 4 h	1	0.52	0.05