Electronic supplementary information (ESI)

Monovalent salt and pH-induced gelation of oxidised cellulose nanofibrils and starch networks: combining rheology and small-angle X-ray scattering

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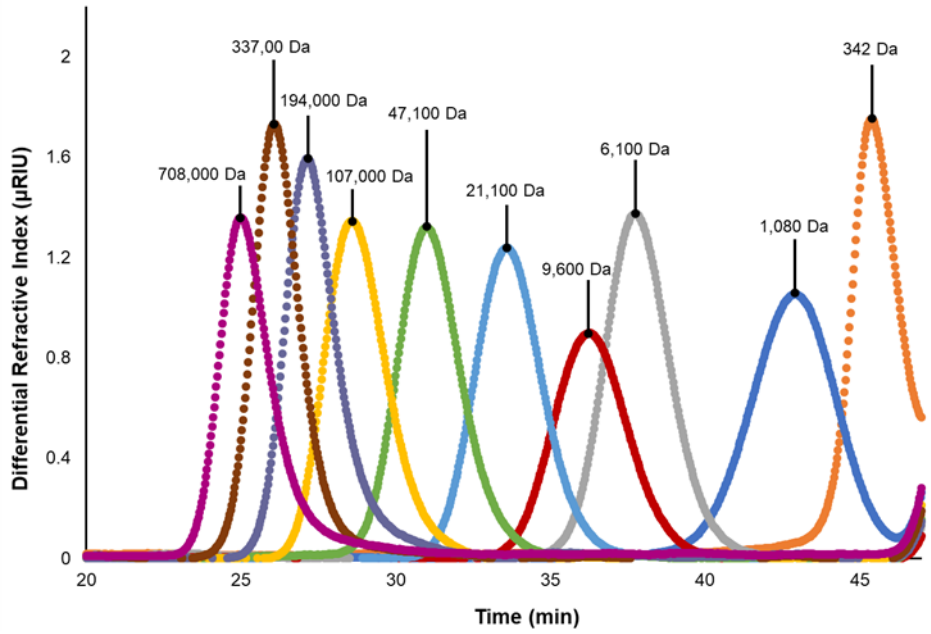
a)



b)

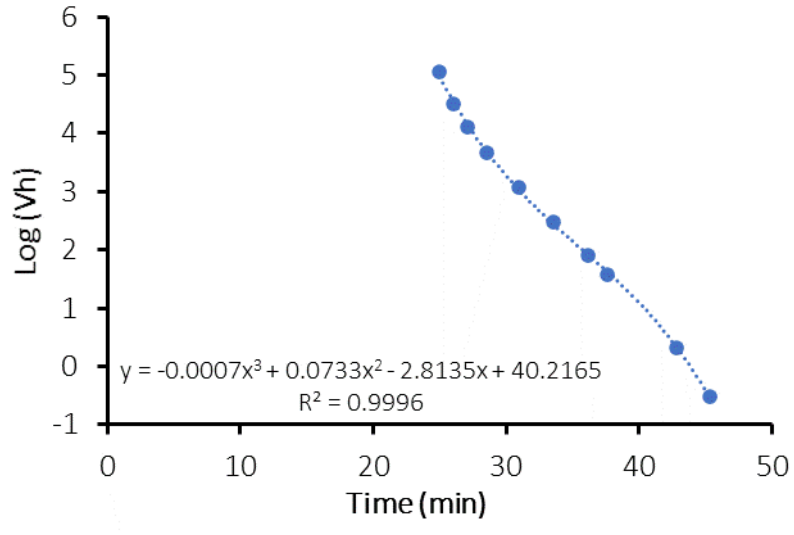


Figure S1: a) Elution profiles of pullulan standards and **b)** standard curve obtained from the elution profile of pullulan standards.

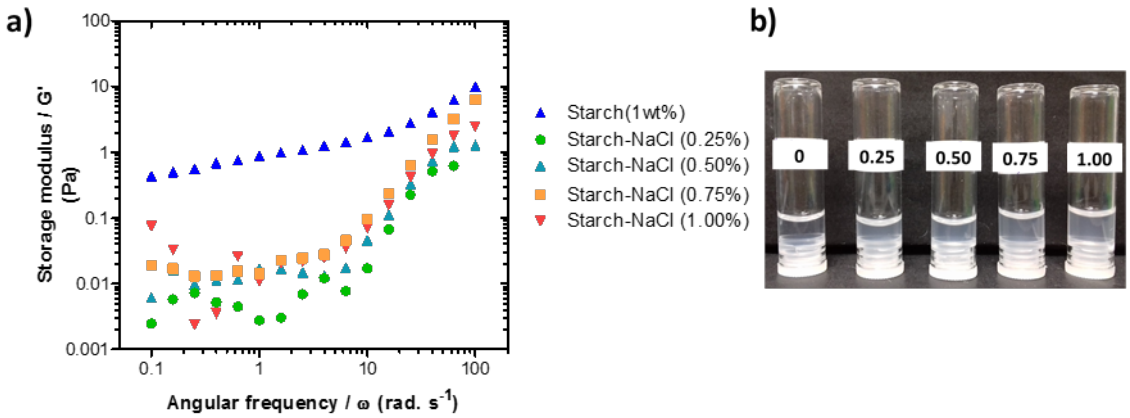


Figure S2: a) Frequency sweep curves and b) photographs of the starch (1 wt%)/NaCl (0, 0.25, 0.5, 0.75 or 1 wt%) hydrogels.

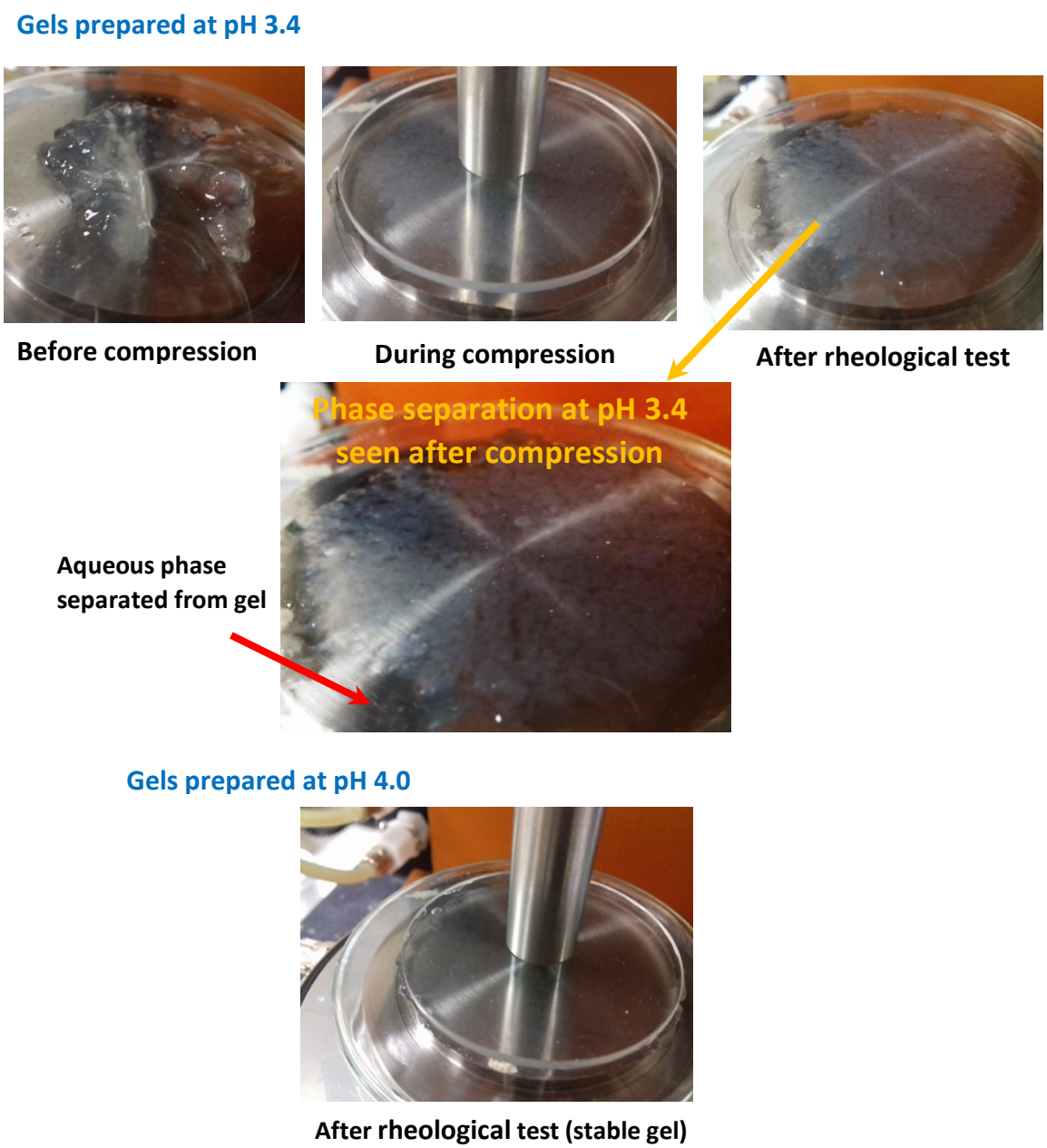


Figure S3: Photographs show the phase separation of OCNF (1 wt%) hydrogels after rheological test produced at pH 3.4 (top) and no phase separation for the gels produced at pH 4.0 (bottom). Here, a 80 mm glass plate was used to visualise the gels physical properties during rheological tests.

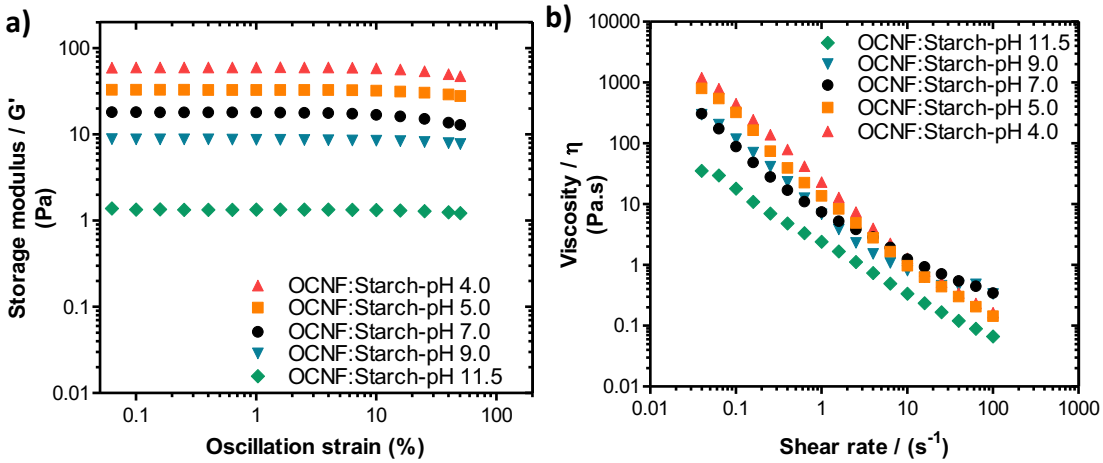


Figure S4: a) amplitude sweeps, and b) shear flow curves of OCNF:Starch (1:1 wt%) hydrogels made at various pH conditions (4, 5, 7, 9, 11.5).

Table S1: The parameters obtained by fitting of SAXS data via rigid and flexible elliptical cylinder models. The length of the OCNF was fixed at 1600 Å based on the average length obtained via the TEM image analysis,[26] except for the OCNF(1%)-NaCl(1%) gel, where a longer length (4000 Å) was used to fit the experimental data. This apparent increase in length might be associated with the salt-induced (at higher concentration, 1%) longitudinally assembled fibres.

SLD (scattering length density) for cellulose: 13.5 x10^{-6} Å^{-2}

SLD solvent: 9.5 x10^{-6} Å^{-2}

	Model	Radius-minor (Å)	Axis ratio	Kuhn length (Å)	Length (Å)
OCNF(1%)	Rigid elliptical cylinder	11±2	4.8	-	1600
OCNF(1%)-0.25% NaCl	Rigid elliptical cylinder	11.5±2	5.2	-	1600
OCNF(1%)-0.50% NaCl	Flexible elliptical cylinder	11.8±2	5.0	600	1600
OCNF(1%)-1.00% NaCl	Flexible elliptical cylinder	12.5±2	5.0	400	4000
OCNF(1%)-pH 4.0	Rigid elliptical cylinder	11.5±2	4.8	-	1600
OCNF(1%)-pH 5.0	Rigid elliptical cylinder	11.5±2	4.8	-	1600
OCNF(1%)-pH 9.0	Rigid elliptical cylinder	11.5±2	4.5	-	1600
OCNF(1%)-pH 11.5	Rigid elliptical cylinder	12.5±2	4.5	-	1600