Electronic supplementary information ESI:

Nanoparticle Rearrangement Under Stress in Cellulose Nanofibrils Networks using *in situ* SAXS During Tensile Testing

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Ex Situ Tensile Testing

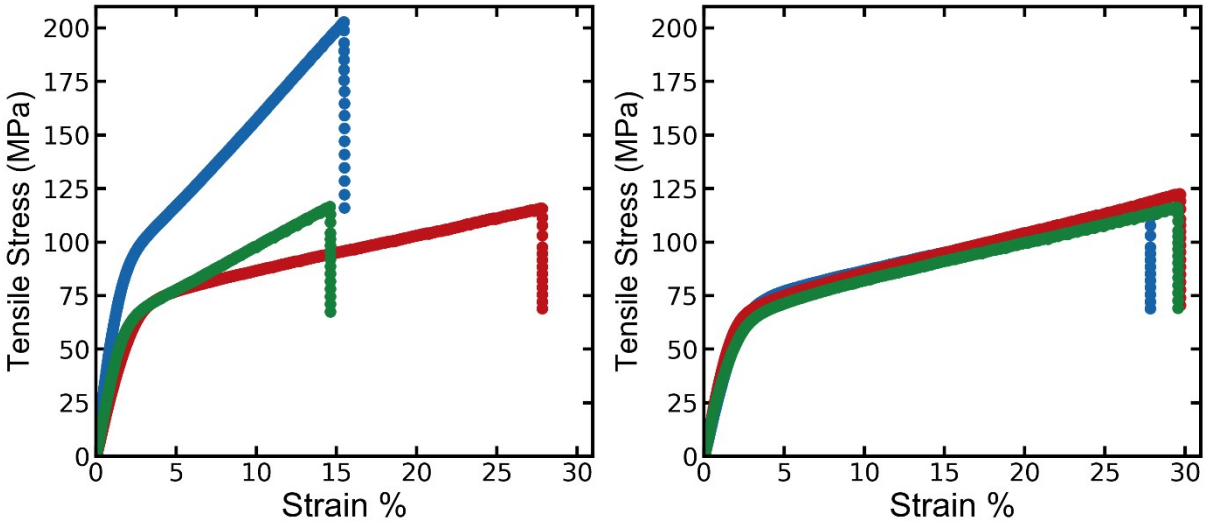

Figure S1: Representative curves from *ex situ* tensile testing of (A) CNF reference (blue), CNF-$PMMA_{100}$ (red), and CNF-$PBMA_{82}$ (green). (B) Three consecutive tests of CNF-$PMMA_{100}$.

SEM of CNF-PBMA$_{82}$

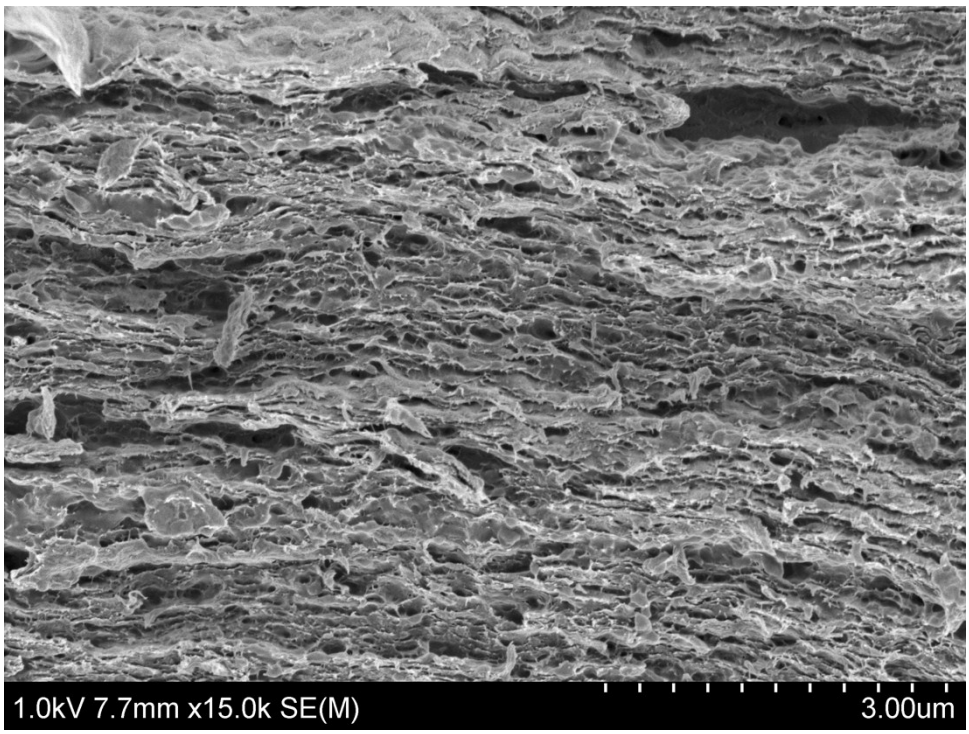

Figure S2: SEM image of cross-section from tensile testing of a composite CNF-PBMA$_{82}$ treated only at room temperature.

Form Factor SAXS

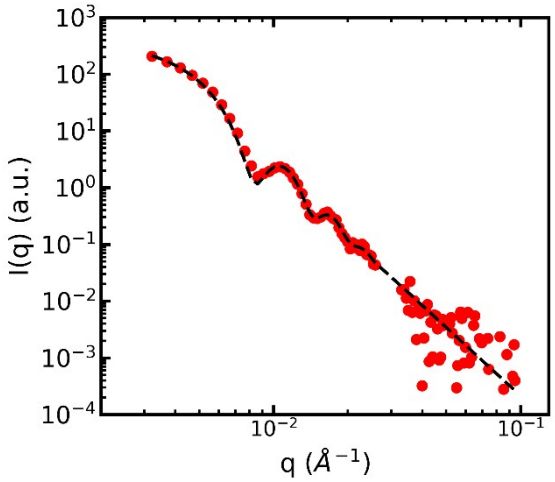

Figure S3: SAXS form factor of $PMMA_{100}$ NPs in dilute aqueous solution. SAXS was performed on the Columbia SAXSLAB equipment and background subtracted. The pattern was fit with a spherical form factor with a radius of 52 nm and a polydispersity of 0.09.

SAXS on 40nm NP Composites

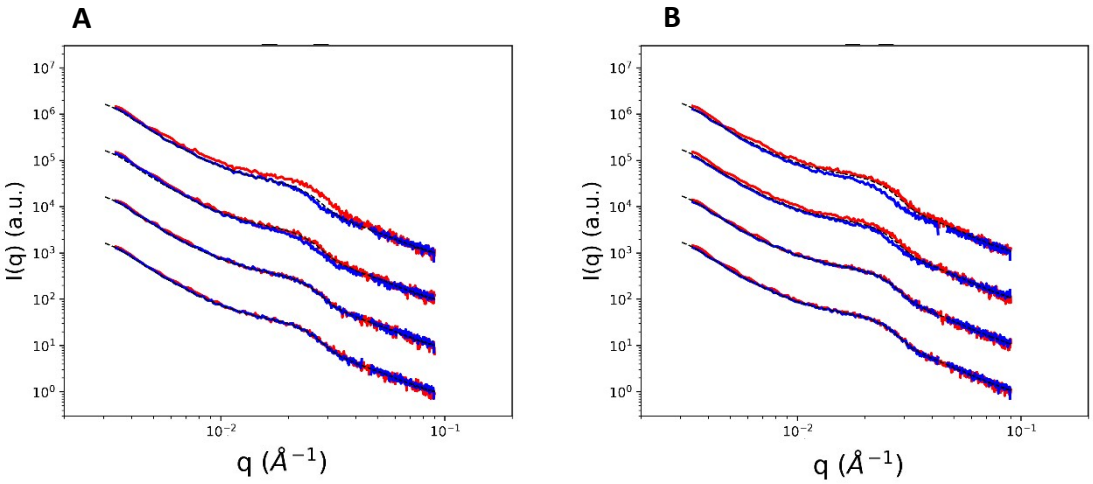

Figure S4: SAXS performed on the (A) CNF-PMMA$_{40}$ and (B) CNF-PBMA$_{40}$ composites during in situ strain at 20% RH at 0, 25, 50, and 95% strain before failure for increasing intensities (2D SAXS curved integrated (blue) vertically and (red) horizontally integrated. Just before fracture, a small degree of anisotropy can be seen around the form factor range of the NPs. The low q range, however, maintains the upturn in intensity with a fractal dimension close to 3 with no apparent anisotropy along this length scale.

Ultra-Small Angle X-ray Scattering

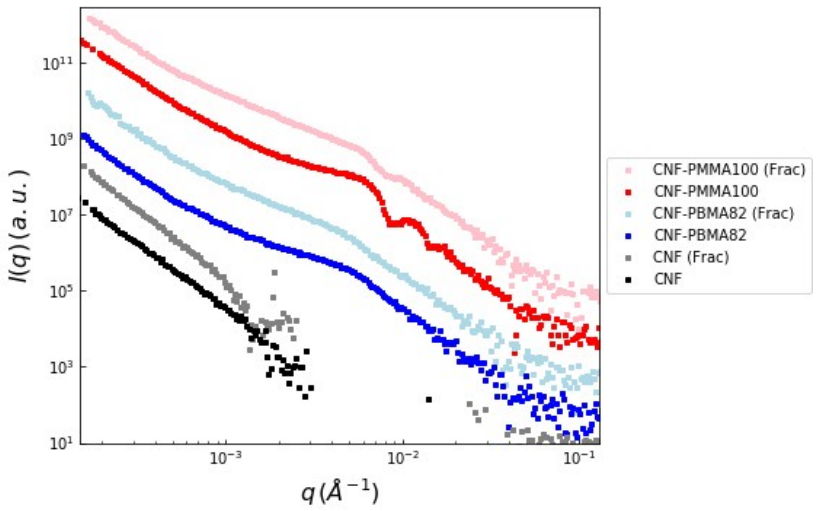

Figure S5: Ultra small angle x-ray scattering performed on CNF, CNF-$PMMA_{100}$, and CNF-$PBMA_{82}$ before and after strain to failure ("Frac"). The smearing of the pattern in the form factor region of the sample is similar to that seen in the *in situ* scattering. The lack of structural anisotropy at low q is noted, down to ~4 micron, for this radially averaged measurement.

Wide Angle X-ray Scattering

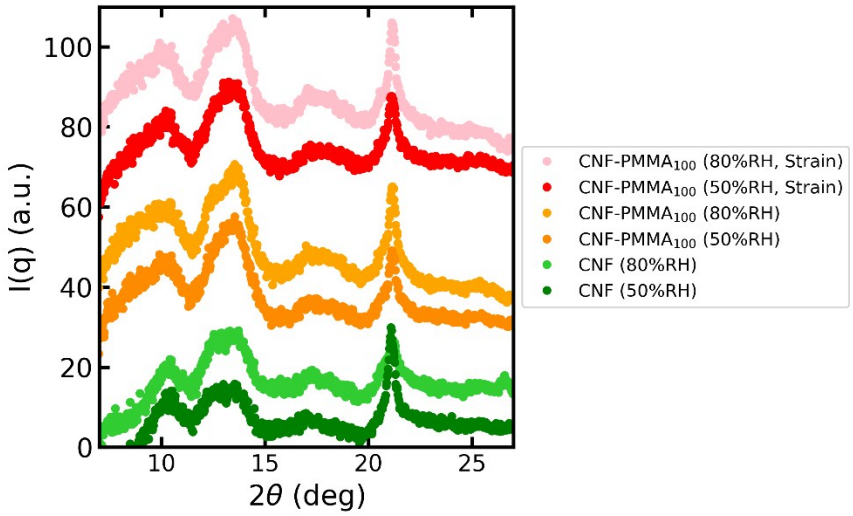

Figure S6: Wide angle X-ray scattering performed on CNF and CNF-$PMMA_{100}$ at different humidities and under in situ strain.

SAXS Fits

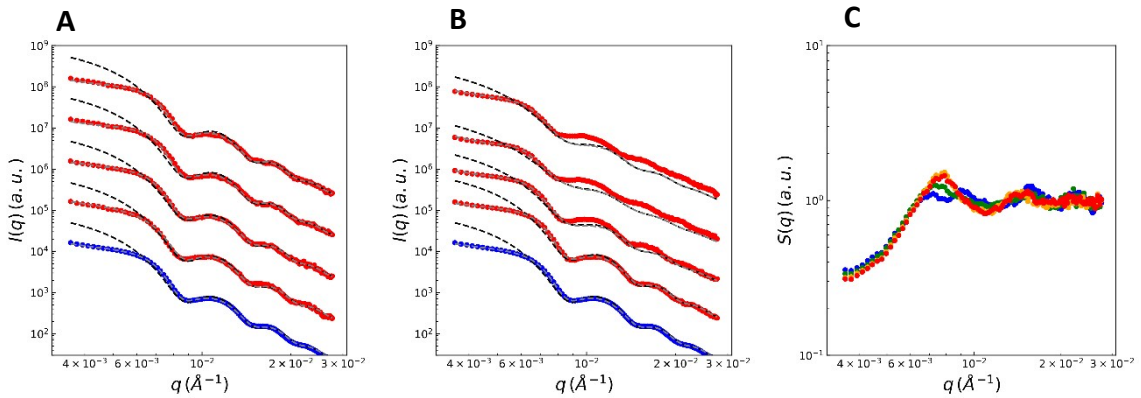

Figure S7: Fitting of the SAXS data for CNF-PMMA$_{100}$ sample for in situ strain at 50% humidity integrated (A) vertically and (B) horizontally. Blue curves are integrated isotropically. Black dashed lines are form factor fits of the NPs. Grey dashed lines are fits incorporating a Percus-Yevick structure factor. Dividing the data in (A) by the form factor gives (C) the structure factor of the NP scattering.