Electronic Supplementary Material (ESI) for Materials Advances.

Supporting Information

Dispersion Engineering of Cellulose Nanofibres in Polyols: For Controlled Microstructure of High-performance Polyurethane Insulation Foam

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S.1 CNF/polyurethane Resin Properties

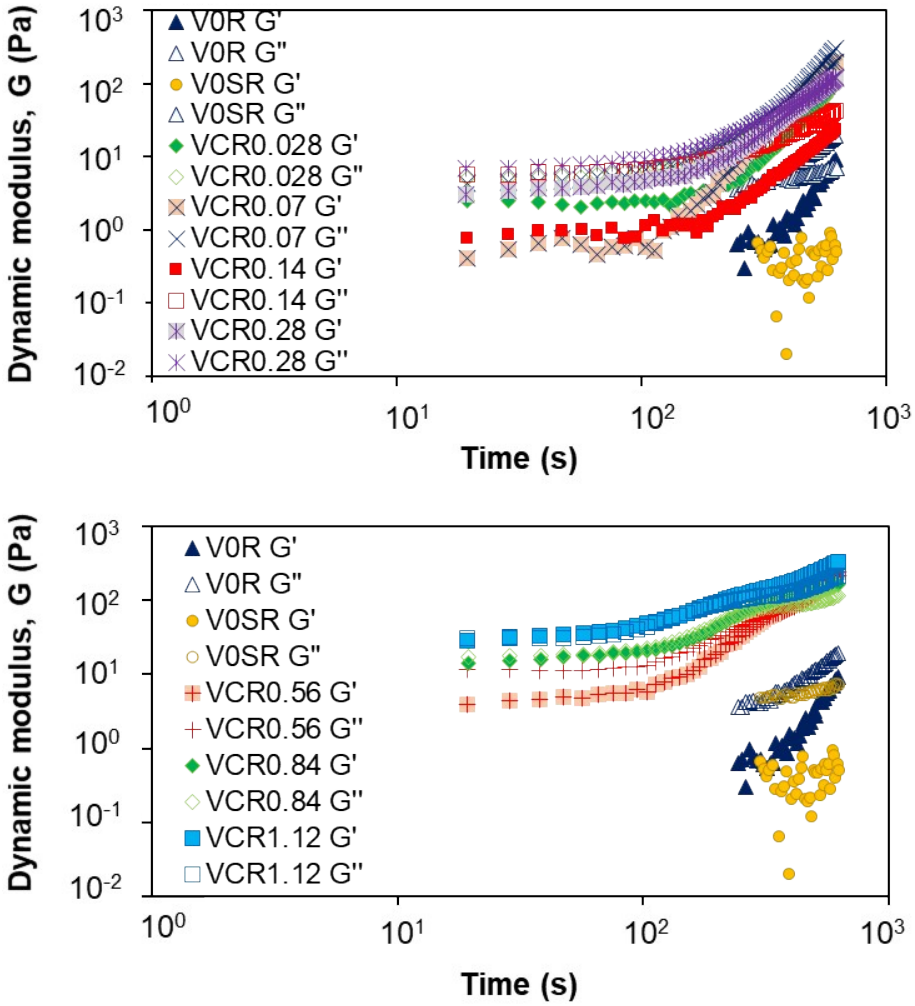

Figure S.1. Dynamic modulus of CNF/polyurethane resins a) at low loadings and b) high loadings (the graph includes mixing time).

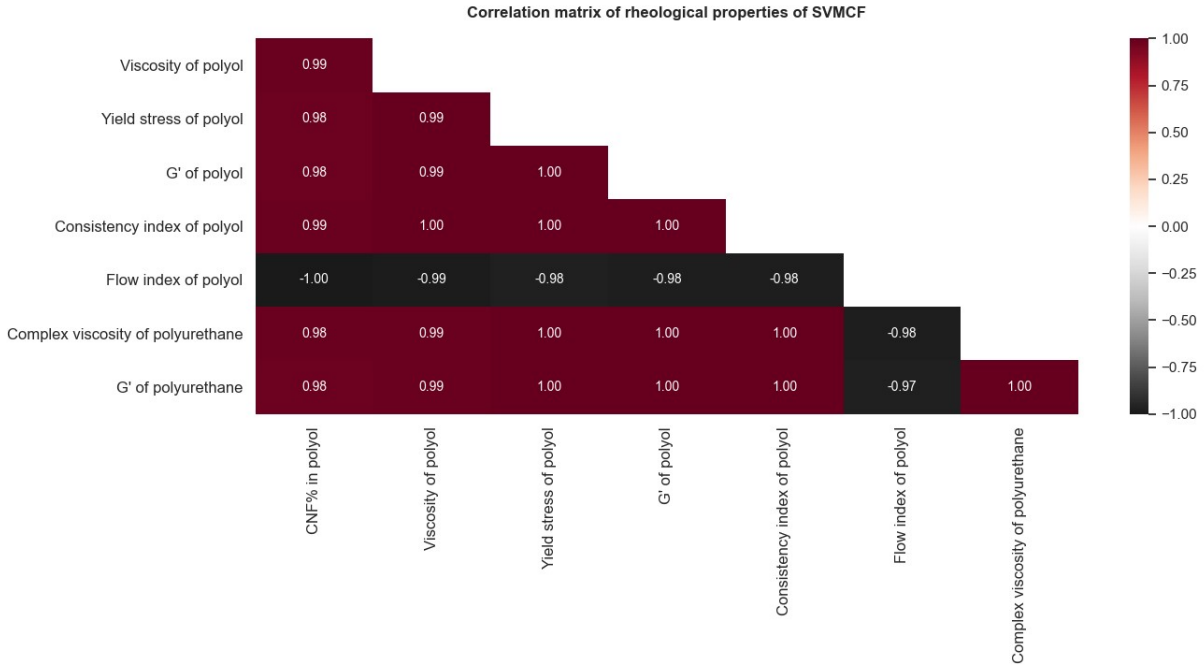

Figure S.2 Correlation chart of rheological properties of the CNF/polyol dispersions and CNF/polyurethane resins

S.2 Chemical Aspects of RPUF Samples

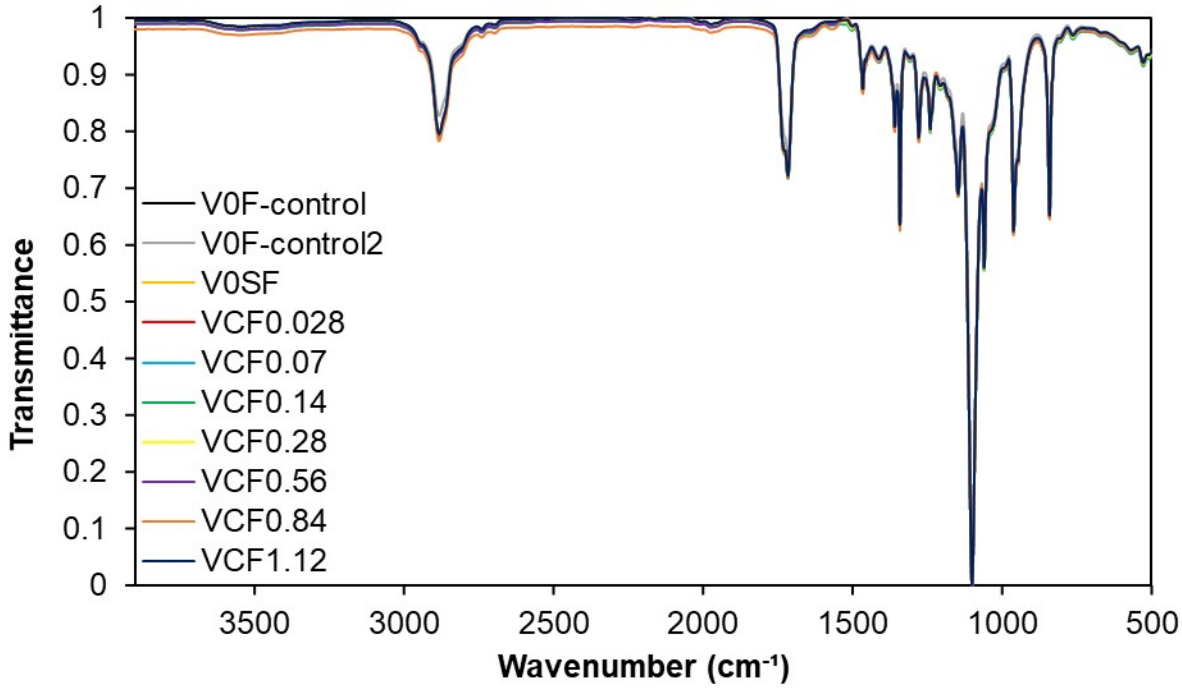

Figure S.3 FTIR-ATR spectra of RPUF samples

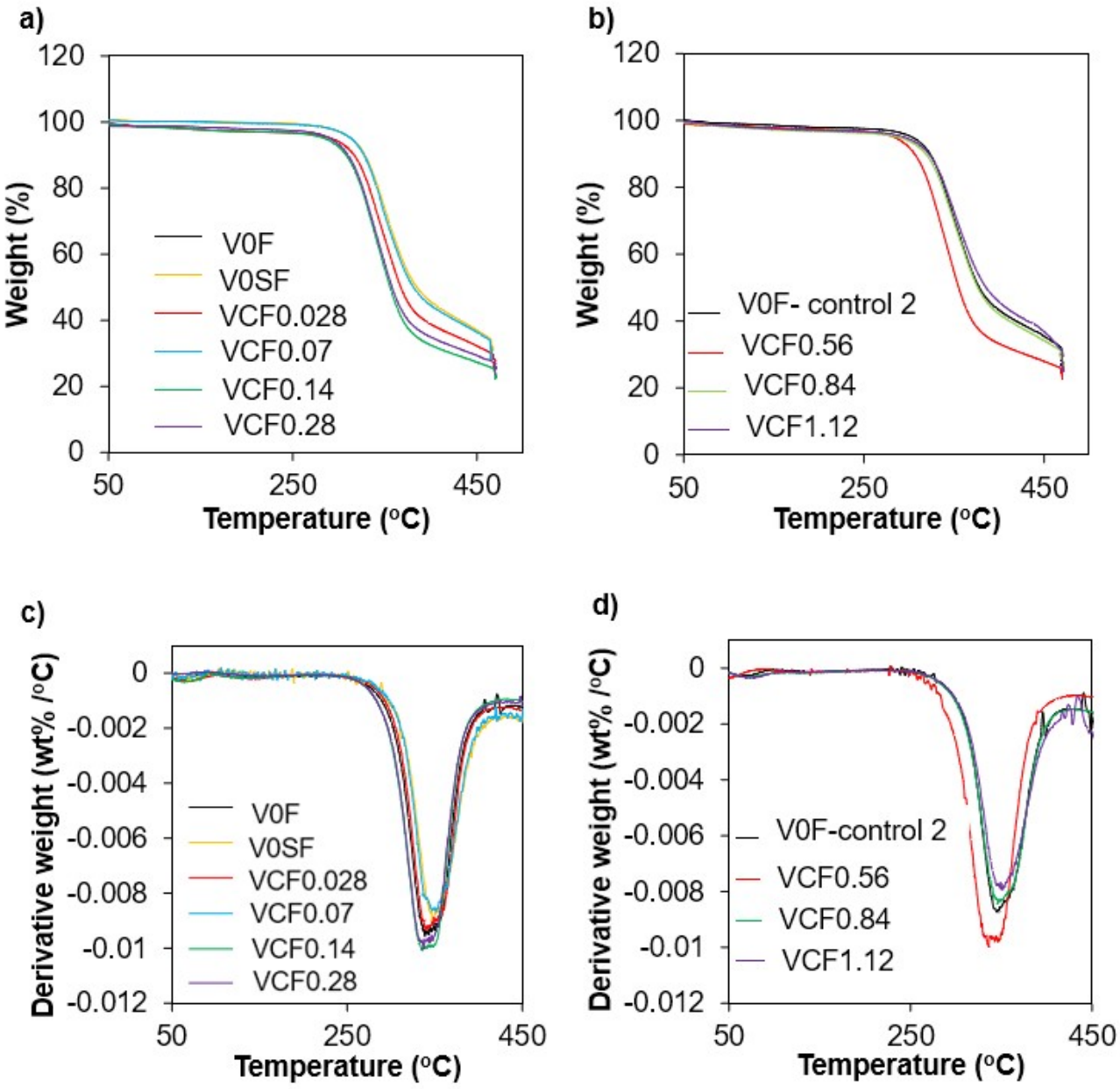

Figure S.4 Effect of the incorporation of CNF in RPUF on thermal Stability for low-density and high-density CNF/RPUF.

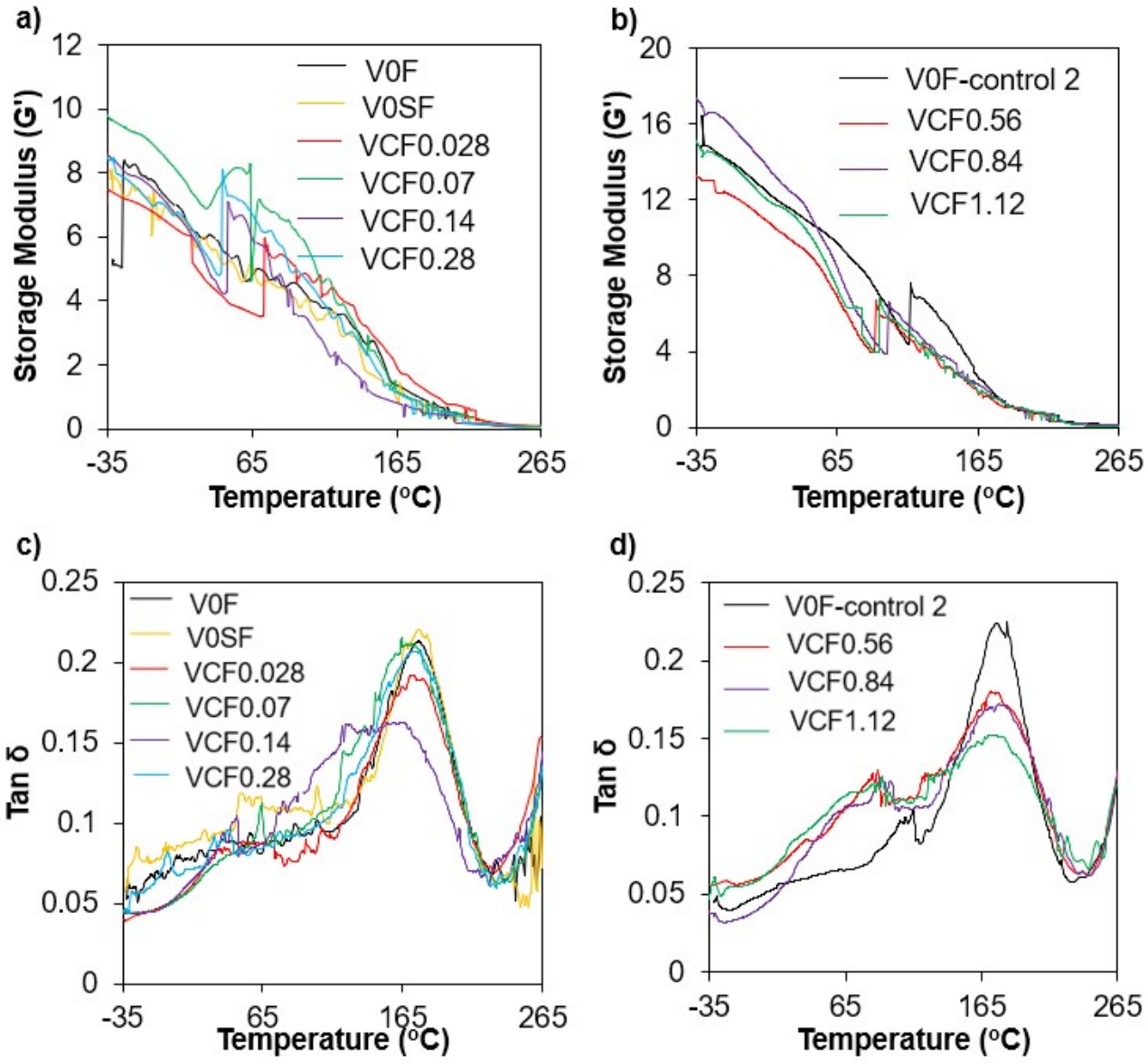

Figure S.5 Effect of incorporation of CNF in RPUF on thermomechanical properties a), b) storage modulus and c), d) tan δ of low-density and high-density CNF/RPUF.

Table S.1 Hydroxyl value analysis (OHv) of CNF/Polyol Dispersions

CNF in polyol (w/w%)	0	0*	0.028	0.07	0.14	0.28	0.56	0.84	1.12
OHv	448 ± 1.4	450 ± 2.5	451 ± 5.2	450 ± 5.2	451 ± 6.3	453 ± 2.5	453 ± 5.2	453 ± 7.6	452 ± 5.2

*Control polyol subjected under high shear

S.3 Microstructural Properties of RPUF Samples

Key: YES = Significant difference, NO = Insignificant difference

Colour code: Green = Significantly improved, Red = Significantly affected

Table S.2 Statistical significance of average cell size of low-density and high-density RPUF samples measured using ImageJ from SEM images (n=3 images, number of min-feret measurements= ~25- 75 for all samples) compared to V0F-control and V0F-control 2

Sample	Change (%)	Significance	Change (%)	Significance
	Parallel		Perpendicular	
Low-density RPUF				
V0F- control				
V0SF	-15.0	YES	-21.2	YES
VCF0.028	-54.0	YES	-47.0	YES
VCF0.07	-44.6	YES	-33.7	YES
VCF0.14	-52.6	YES	-46.5	YES
VCF0.28	-45.8	YES	-38.6	YES
High-density RPUF				
V0F-control 2				
VCF0.56	-31.4	YES	-16.9	YES
VCF0.84	-30.6	YES	-18.6	YES
VCF1.12	-26.4	YES	16.0	YES

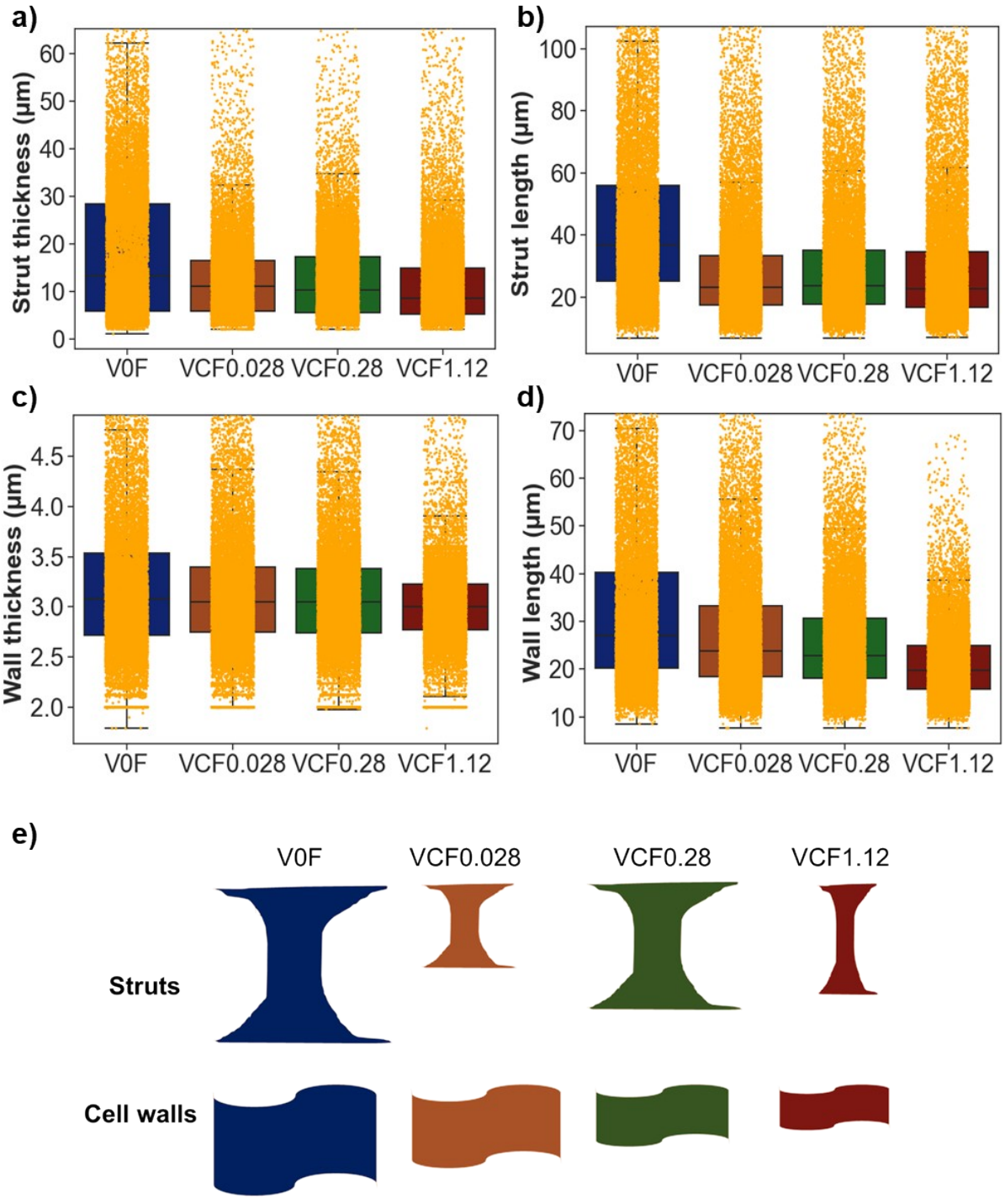

Figure S.6 The interquartile distribution of a) strut thickness, b) wall thickness, c) strut length, and d) wall length of CNF/RPUF; and e) the schematic representation of possible changes in strut and cell wall upon CNF incorporation based on average values

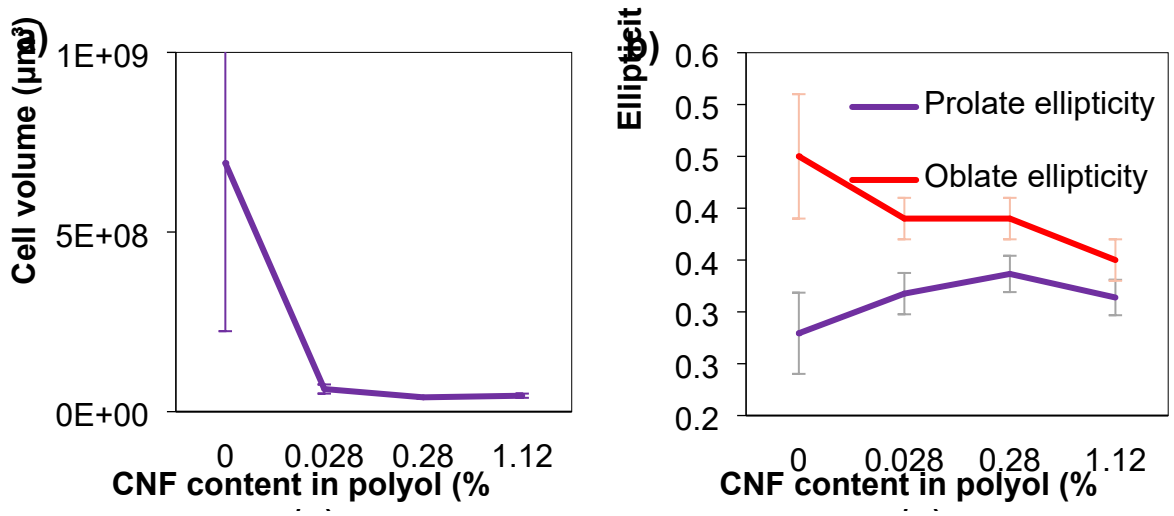

Figure S.7 Influence of CNF loading on spatial microstructural parameters of RPUF sample

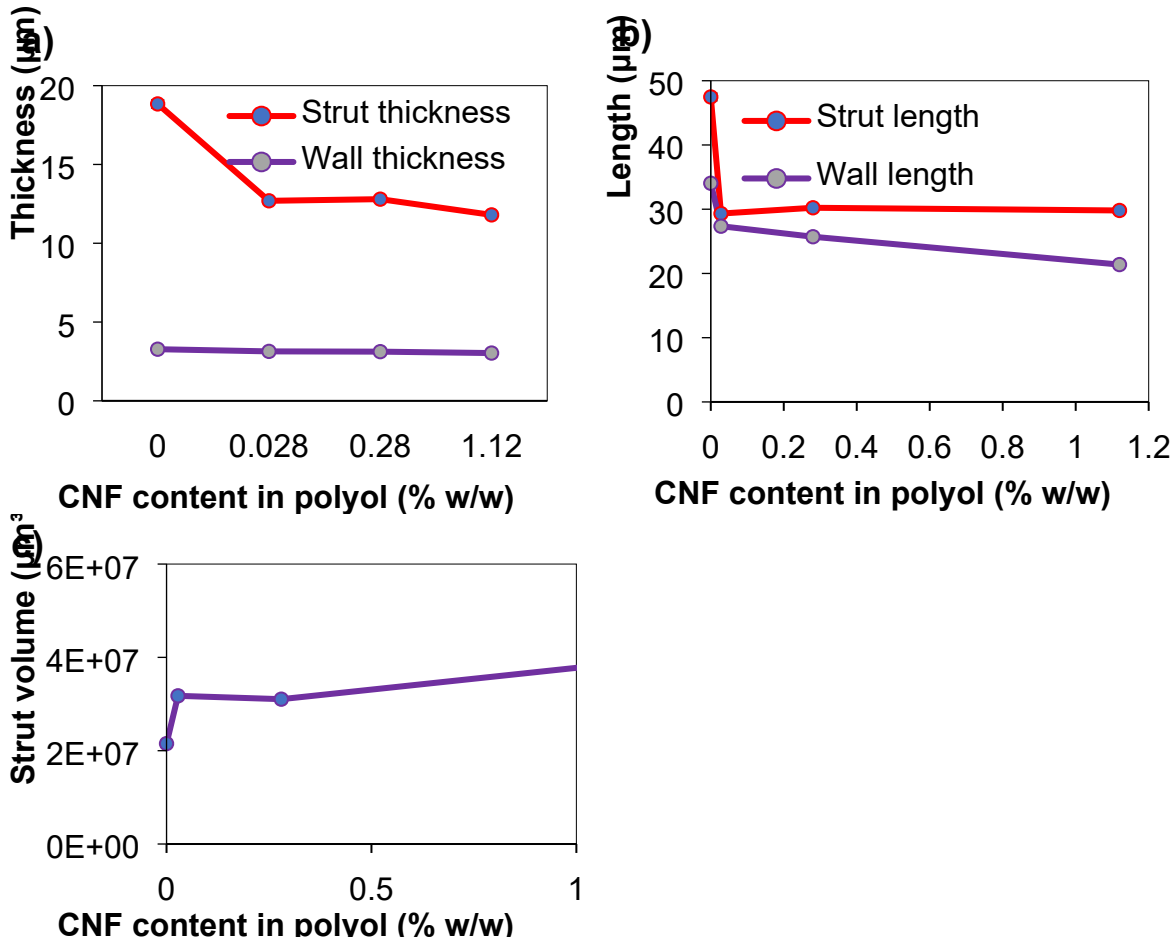

Figure S.8 Influence of CNF loading on skeletal microstructural parameters of RPUF sample

Table S.3 Statistical significance of spatial parameters measured from 3D reconstruction analysis compared to V0F-control (n= 46 to 300)

Properties	Number of cells	Cell volume		Prolate ellipticity		Oblate ellipticity		Sphericity	
		Change (%)	Significance	Change (%)	Significance	Change (%)	Significance	Change (%)	Significance
V0F-control	45								
VCF0.028	190	-90.9	YES	13.7	NO	-13	YES	4.9	YES
VCF0.28	187	-94.2	YES	20.5	YES	-13.4	YES	-0.6	NO
VCF1.12	295	-93.6	YES	12.4	NO	-21.5	YES	3.6	NO

Table S.4 Significance of Spatial Parameters Measured from 3D reconstruction analysis compared to CNF/RPUF (n= 46 to 300)

Comparison sample	Sample	Cell volume	Sphericity	Prolate ellipticity	Oblate ellipticity
VCF0.028	VCF0.28	NO	NO	NO	YES
	VCF1.12	NO	NO	NO	YES
VCF0.28	VCF1.12	YES	NO	NO	YES

Table S.5 Significance of Skeletal Parameters Measured from 2D reconstruction analysis compared to V0F-control (n= 16001 to 260000)

Properties	Strut length		Strut thickness		Wall length		Wall thickness	
	Change (%)	Significance	Change (%)	Significance	Change (%)	Significance	Change (%)	Significance
V0F- control								
VCF0.028	-38.2	YES	-32.6	YES	-19.6	YES	-4.2	YES
VCF0.28	-36.3	YES	-32.1	YES	-24.4	YES	-4.7	YES
VCF1.12	-37.2	YES	-37.4	YES	-37.2	YES	-7.5	YES

Table S.6 Significance of Skeletal Parameters Measured from 2D reconstruction analysis compared to CNF/RPUF (n= 160001 to 260000)

Comparison sample	Sample	Strut thickness	Strut length	Wall length	Wall thickness
VCF0.028	VCF0.28	NO	YES	YES	NO
	VCF1.12	NO	YES	YES	NO
VCF0.28	VCF1.12	NO	NO	YES	NO

S.4 Statistical Significance of Physical Properties of RPUF Samples

Table S.7 Statistical significance of compressive properties of low-density and high-density RPUF compared to V0F-control and V0F-control 2 respectively (n=6)

	Density		CS		Sp. CS		CM		Sp. CM	
	Change (%)	**Significance**	**Change (%)**	**Significance**	**Change (%)**	**Significance**	**Change (%)**	**Significance**	**Change (%)**	**Significance**
Low-density RPUF -Parallel										
V0F- control										
V0SF	-5.6	YES	-16.8	YES	-12.0	NO	-10.9	NO	-5.8	NO
VCF0.028	19.3	YES	2.0	NO	-14.7	NO	11.0	NO	-7.2	NO
VCF0.07	16.3	YES	9.9	NO	-5.6	NO	28.3	YES	10.2	NO
VCF0.14	22.4	YES	11.9	NO	-8.7	NO	29.6	NO	5.8	NO
VCF0.28	21.4	YES	12.9	NO	-6.9	NO	32.2	YES	16.4	NO
Low-density RPUF -Perpendicular										
V0F- control										
V0SF	-1.55	NO	-3.9	NO	-1.3	NO	10.0	NO	12.9	NO
VCF0.028	20.23	YES	50.6	YES	25.2	YES	88.2	YES	56.5	YES
VCF0.07	16.67	YES	42.9	YES	22.4	YES	64.8	YES	41.3	YES
VCF0.14	23.54	YES	46.8	YES	18.7	YES	74.7	YES	41.4	YES
VCF0.28	24.38	YES	49.4	YES	20.0	YES	67.3	YES	34.5	YES
High-density RPUF -Parallel										
V0F-control										

2										
VCF0.56	-6.4	YES	-24.7	YES	-19.4	YES	-21.1	NO	-15.6	NO
VCF0.84	11.2	YES	-14.9	NO	-23.4	YES	-13.3	NO	-21.9	NO
VCF1.12	12.4	YES	-16.7	NO	-25.9	YES	-17.3	NO	-26.5	NO
High-density RPUF -Perpendicular										
V0F-control 2										
VCF0.56	-4.4	YES	-31.6	YES	-28.5	YES	-31.4	YES	-28.2	YES
VCF0.84	14.2	YES	-17.7	YES	-27.9	YES	-16.6	YES	-27.0	YES
VCF1.12	13.2	YES	-13.4	YES	-23.5	YES	-10.8	YES	-21.2	YES

Footnotes: CS= compressive strength, Sp. CS= Specific compressive strength, CM= compressive modulus, Sp. CM= Specific compressive modulus

Table S.8 Statistical significance of compressive properties of low-density RPUF samples compared to V0SF (n=6)

	Density		CS		Sp. CS		CM		Sp. CM	
	Change (%)	**Significance**	**Change (%)**	**Significance**	**Change (%)**	**Significance**	**Change (%)**	**Significance**	**Change (%)**	**Significance**
Low-density RPUF -Parallel										
V0F-control	6.0	YES	20.2	YES	13.6	NO	12.2	NO	6.1	NO
V0SF										
VCF0.028	26.4	YES	22.6	NO	-3.1	NO	24.5	NO	-1.5	NO
VCF0.07	23.2	YES	32.1	YES	7.2	NO	44.0	YES	17.0	NO
VCF0.14	29.7	YES	34.5	YES	3.7	NO	45.4	YES	12.2	NO
VCF0.28	28.6	YES	35.7	YES	5.7	NO	48.3	YES	23.5	NO
Low-density RPUF -Perpendicular										
V0F-control	1.6	NO	4.1	NO	1.3	NO	-9.1	NO	-11.4	NO
V0SF										
VCF0.028	22.1	YES	56.8	YES	26.9	YES	71.2	YES	38.6	YES
VCF0.07	18.5	YES	48.6	YES	24.0	YES	49.8	YES	25.1	YES
VCF0.14	25.5	YES	52.7	YES	20.3	NO	58.9	YES	25.2	YES
VCF0.28	26.3	YES	55.4	YES	21.6	NO	52.1	YES	19.1	NO

Footnotes: CS= compressive strength, Sp. CS= Specific compressive strength, CM= compressive modulus, Sp. CM= Specific compressive modulus

Table S.9 Statistical significance of thermal conductivity of low- density and high density RPUF samples (n=3) in comparison to V0F- control and V0F- control 2 respectively at 1 month and 6 months

Sample in comparison with control	**Significance**	**Percentage of change**	**Significance**	**Percentage of change**
Months	1		6	
Low-density RPUF				
V0F- control				
V0SF	YES	16	YES	14
VCF0.028	YES	-20	YES	-15
VCF0.07	NO	-5	NO	-2
VCF0.14	YES	-19	YES	-17
VCF0.28	YES	-10	NO	-6
High-density RPUF				
V0F-control 2				
VCF0.56	YES	-18	NO	-16
VCF0.84	NO	-17	NO	-15
VCF1.12	NO	-16	NO	-14

Table S.10 Statistical significance of thermal conductivity of low- density RPUF samples (n=3) in comparison to V0SF respectively at 1 month and 6 months

Sample in comparison with V0SF	Significance	Percentage of change	Significance	Percentage of change
Months	1		6	
Low-density RPUF				
V0F- control	-14	YES	-12	YES
V0SF				
VCF0.028	-31	YES	-26	YES
VCF0.07	-18	NO	-14	NO
VCF0.14	-30	YES	-28	YES
VCF0.28	-22	YES	-18	NO

Table S.11 Statistical significance of thermal conductivity of RPUF samples (n=3) comparison before and after ageing

Sample	Percentage of change	Significance
Low-density RPUF		
V0F- control	7.25	YES
V0SF	5.52	NO
VCF0.028	12.81	NO
VCF0.07	10.71	NO
VCF0.14	9.68	NO
VCF0.28	12.1116	NO
High-density RPUF		
V0F-control 2	6.37321	NO
VCF0.56	9.3715	YES
VCF0.84	9.5328	YES
VCF1.12	9.9088	YES

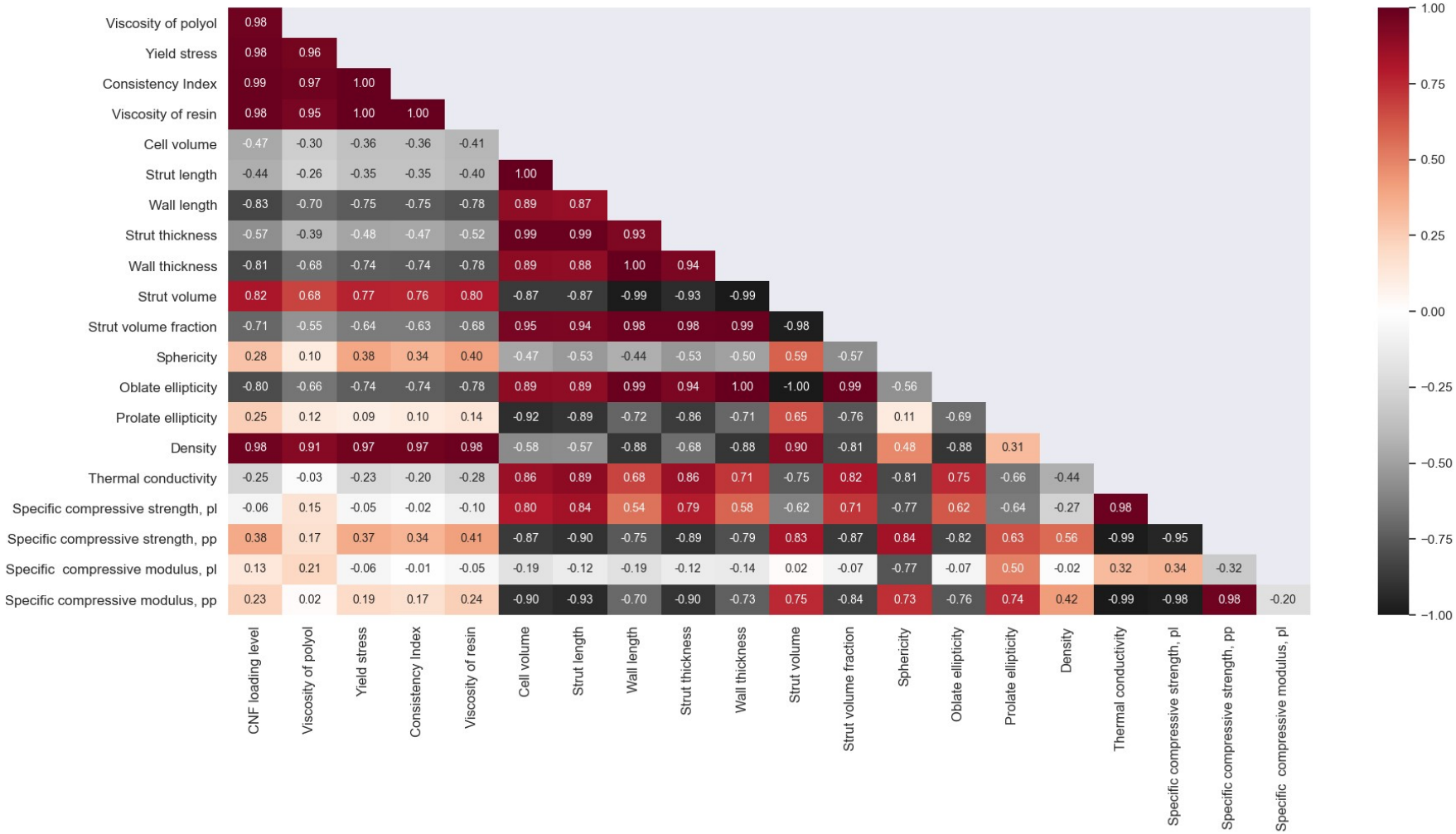

Figure S.9 Correlation of rheological properties of CNF/polyol dispersions and CNF/polyurethane resins with the microstructural and physical properties of the CNF/RPUF

Sample	V0F-control	V0SF	VCF0.028	VCF0.07	VCF0.14	VCF0.28	V0F-control 2	VCF0.56	VCF0.84	VCF1.12
Properties										
Core density (kg/m^3)	32.7 ± 2.3	31.0 ± 2.4	39.3 ± 3.1	37.9 ± 2.2	39.5 ± 2.0	40.5 ± 4.3	55.6 ± 1.8	53.2 ± 3.5	62.3 ± 2.7	63.1 ± 3.0
Average cell size diameter (μm) (‖)*	669 ± 59	569 ± 57	308 ± 30	371 ± 15	317 ± 11	362 ± 15	453 ± 24	311 ± 14	314 ± 15	334 ± 17
Average cell size diameter (μm) (⊥)*	663 ± 75	522 ± 26	351 ± 21	440 ± 27	355 ± 13	407 ± 19	415 ± 26	345 ± 16	337 ± 18	481 ± 28

*Data from SEM, λ = Thermal conductivity, ‖ = parallel to rise, ⊥ = perpendicular to rise, and RT = room temperature.

S.5 Microstructural parameters of CNF/RPUF samples