Effects of different moisture content on structural and functional properties cellulose with cell wall components in different citrus fibres.

Kaja Kristensen[1], George Warne[1], Deepa Agarwal*[1,2], Tim J. Foster[1]

1 Division of Food, Nutrition, and Dietetics, School of Biosciences, University of Nottingham, Sutton Bonington Campus, Loughborough, LE12 5RD, UK.

2 The New Zealand Institute of Plant and Food Research, Plant & Food Research Canterbury Agriculture & Science Centre, Gerald St, Lincoln 7608, New Zealand.

**Corresponding author email: Deepa.Agarwal@plantandfood.co.nz*

Supplementary Data:

Supplementary Figures:

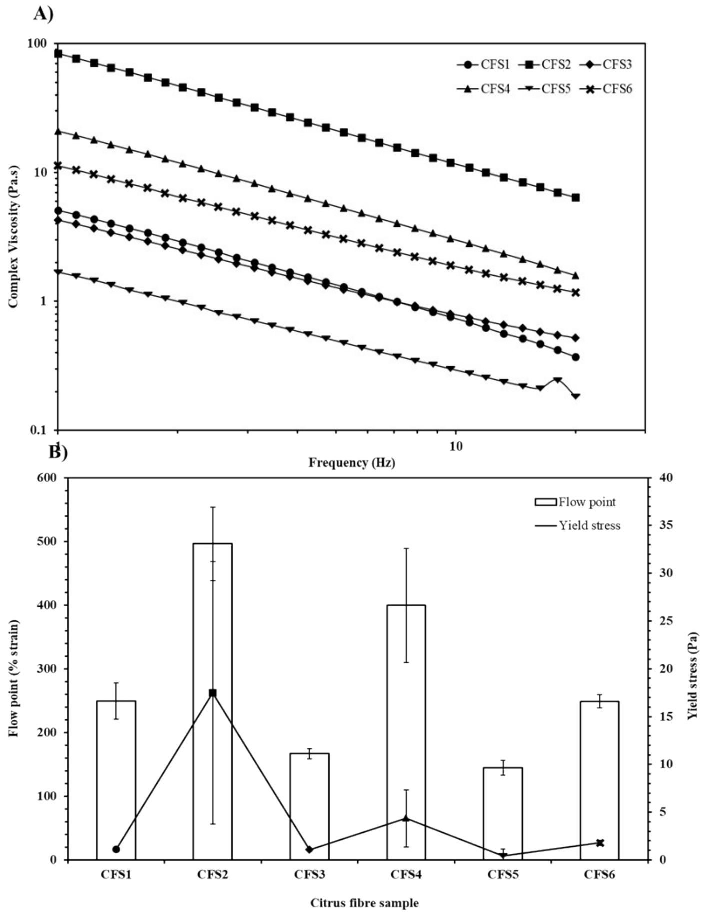
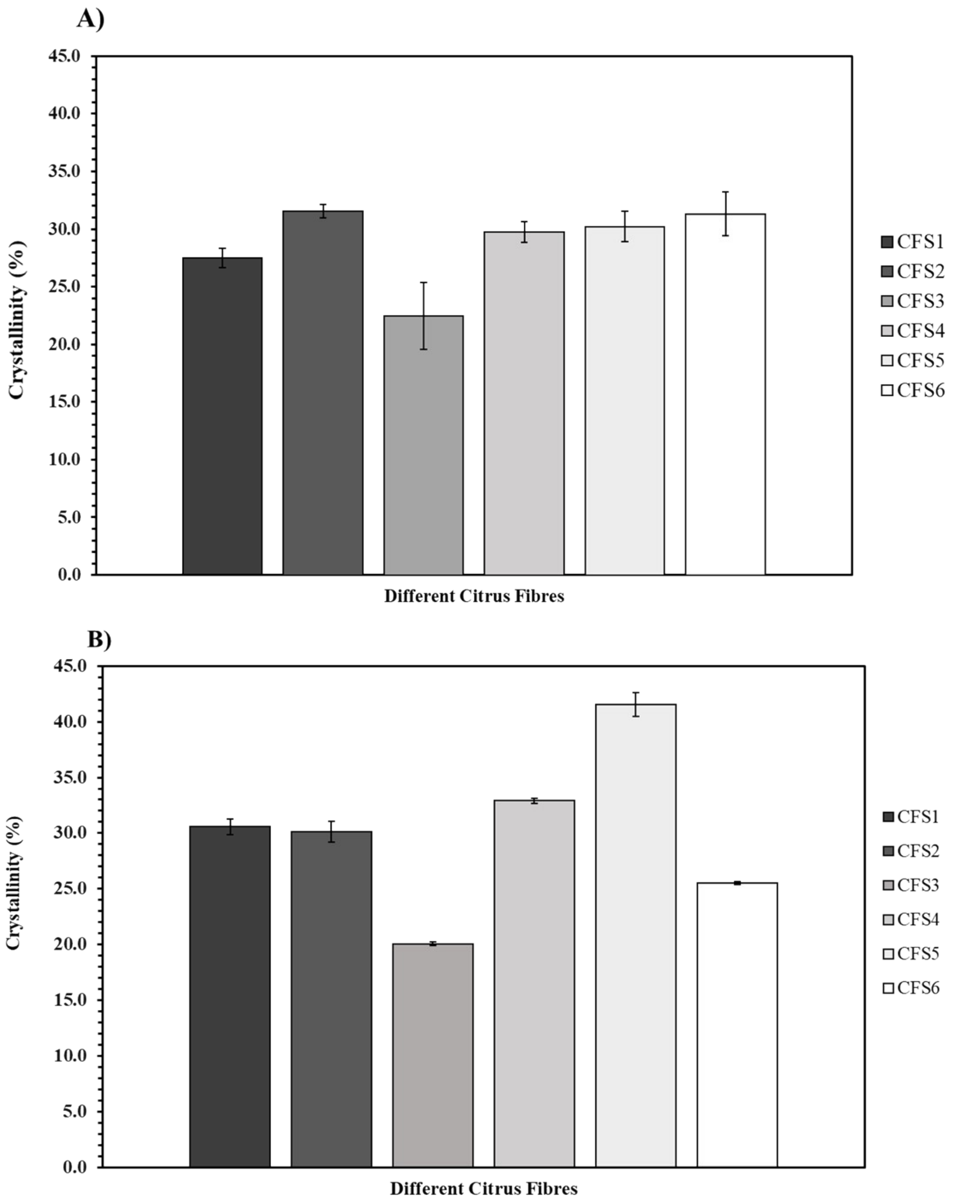
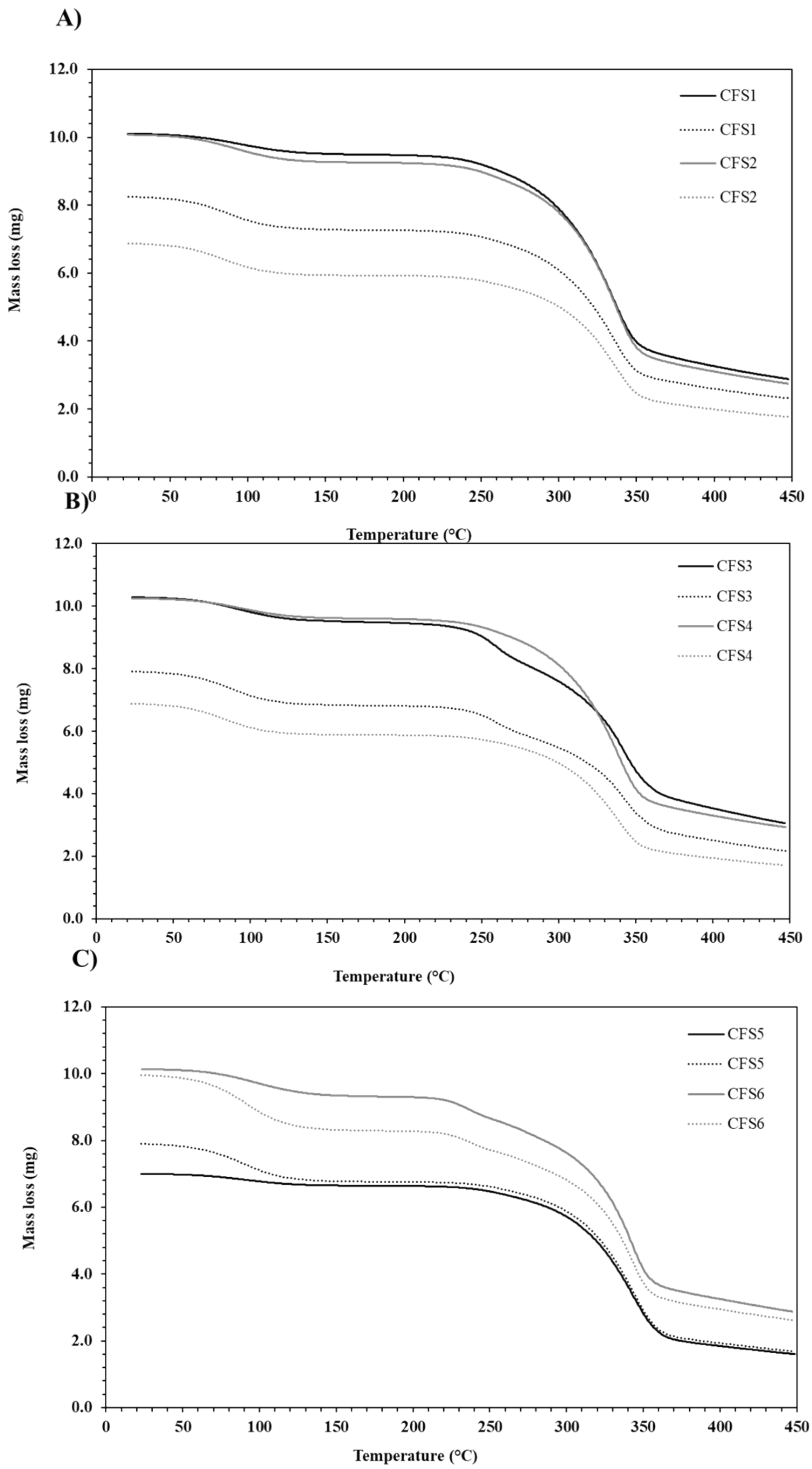


Figure S1. *Rheological properties of 2% w/w aqueous citrus fibre suspensions (CFS1-CFS6), where A) is the complex viscosity measured as a function of frequency at 0.2% shear strain, B) is the flow point and yield stress values taken at 1 Hz constant frequency from the amplitude sweeps. All values were measured at 20±1 °C.*



Supplementary Figure S2. *Degree of crystallinity for the citrus fibre samples (CFS1-CFS6), where A) low moisture and B) medium moisture. The plots are illustrated as the average values of two replicates ± standard deviations.*



Supplementary Figure S3. *Thermogravimetric curves of samples A) CFS1 & CFS2*, *B) CFS3 & CFS4, and C) CFS5 & CFS6*, *where solid lines represent samples with low moisture (5-7% w/w), and dashed lines represent samples with medium moisture (14-17% w/w)*.

Supplementary Tables:

Table S1. *Absorbance band assignment according to Boeriu, Bravo, Gosselink, & van Dam (2004), Carrillo, Colom, Suñol, & Saurina (2004), Célino, Gonçalves, Jacquemin, & Fréor (2014), Olsson & Salmén (2004), and Poletto, Júnior, & Zattera (2014).*

Wave number (cm^{-1})	Assignment	Component
3600-3300	-OH stretching	Carbohydrate, lignin
2900-2855, 1360-1310	-CH, $-CH_2$, $-CH_3$ stretching	Carbohydrate, lignin
1730	C=O stretching	Pectin, hemicellulose
1620	C=O, C=C, -OH stretching	Carbohydrate, lignin
1600, 1500, 1260	Aromatic ring vibration	Lignin
1415	-CH, COO^- vibration	Carbohydrate, lignin
1230	C=C, C=O, C-O vibration	Lignin, pectin, hemicellulose
1150-1020	R-O-R, C-O stretching	Carbohydrate, lignin
890	β-links	Cellulose

Table S2. *Peak areas at 1595, 1509, 1275, and 1220 cm^{-1} for the citrus fibre samples (CFS1-CFS6) in low moisture (LM) and medium moisture (MM) systems. Data are presented as the mean value of five replicates ± standard deviations.*

	Peak area at 1595 cm^{-1}	Peak area at 1509 cm^{-1}	Peak area at 1275 cm^{-1}	Peak area at 1220 cm^{-1}
Lignin[1]	2.61±0.49	1.94±0.45	2.15±0.57	2.83±0.68
CFS1LM	0.91±0.23	0.53±0.19	1.26±0.36	0.81±0.29
CFS1MM	1.64±0.20	0.80±0.13	1.32±0.26	0.66±0.21
CFS2LM	0.72±0.13	0.25±0.09	0.86±0.18	0.43±0.16
CFS2MM	1.27±0.54	0.45±0.26	0.84±0.511	0.29±0.23
CFS3LM	0.82±0.35	0.37±0.27	0.95±0.53	0.77±0.50

CFS3MM	1.88±0.29	0.99±0.17	2.01±0.39	1.61±0.37
CFS4LM	0.57±0.15	0.27±0.12	0.82±0.23	0.43±0.19
CFS4MM	1.20±0.28	0.40±0.21	0.88±0.35	0.29±0.25
CFS5LM	0.42±0.10	0.08±0.06	0.77±0.17	0.35±0.13
CFS5MM	1.30±0.19	0.33±0.11	1.33±0.24	0.49±0.18
CFS6LM	0.67±0.24	0.28±0.17	0.71±0.32	0.59±0.30
CFS6MM	2.19±0.16	0.48±0.12	1.38±0.19	0.80±0.17

[1] Lignin was obtained from Sigma-Aldrich (Gillingham, UK).

Quantification of the peak areas at 1595 (1610-1579), 1509 (1524-1495), 1275 (1290-1260), and 1220 (1234-1206) cm^{-1} was adapted from Huang et al. (2012) (Table S2). Peak areas were baseline corrected using asymmetric least squares regression in OriginPro 9.8.0.200 (OriginLab Corporation, USA). The following parameters were used: Asymmetric factor (0.0001), Threshold (0.005), Smoothing factor (6.5), and Number of iterations (10). Peak areas were identified through OriginPro at these specified wavelengths with ± 15 cm^{-1}.

Table S3. *Loadings of infrared vibration data on PC-1 and PC-2.*

	Positive loadings	Negative loadings
PC-1	2880, 2855, 1450, 1269, 1088	3626, 3453, 3262, 3082, 1651
PC-2	3697, 3120, 1724, 1450, 907	3406, 2829, 1179

Table S4. *Derivative thermogravimetric peak temperatures and mass loss rates for the citrus fibre samples (CFS1-CFS6) in low moisture (LM) and medium moisture (MM) systems.*

	Starting point		Moisture peak		Pectin peak[1]		Cellulose peak[2]	
	Temp. (°C)	Mass loss rate (mg·s^{-1})	Temp. (°C)	Mass loss rate (mg·s^{-1})	Temp. (°C)	Mass loss rate (mg·s^{-1})	Temp. (°C)	Mass loss rate (mg·s^{-1})

CFS1LM	23.0	$9.73\cdot10^{-5}$	93.4	$1.45\cdot10^{-3}$	-	-	336	$1.75\cdot10^{-2}$
CFS1MM	23.1	$1.84\cdot10^{-4}$	86.8	$2.87\cdot10^{-3}$	-	-	336	$1.32\cdot10^{-2}$
CFS2LM	23.0	$1.15\cdot10^{-4}$	92.3	$2.14\cdot10^{-3}$	-	-	337	$1.83\cdot10^{-2}$
CFS2MM	23.1	$2.16\cdot10^{-4}$	84.3	$2.83\cdot10^{-3}$	-	-	338	$1.17\cdot10^{-2}$
CFS3LM	23.4	$1.10\cdot10^{-4}$	92.1	$2.04\cdot10^{-3}$	259	$6.48\cdot10^{-3}$	342	$1.47\cdot10^{-2}$
CFS3MM	23.1	$2.04\cdot10^{-4}$	87.2	$3.17\cdot10^{-3}$	259	$2.87\cdot10^{-3}$	342	$1.08\cdot10^{-2}$
CFS4LM	23.2	$7.53\cdot10^{-5}$	93.2	$1.65\cdot10^{-3}$	-	-	339	$1.82\cdot10^{-2}$
CFS4MM	23.0	$2.66\cdot10^{-4}$	84.0	$3.03\cdot10^{-3}$	-	-	339	$1.14\cdot10^{-2}$
CFS5LM	23.1	$5.54\cdot10^{-5}$	92.1	$9.09\cdot10^{-4}$	-	-	343	$1.38\cdot10^{-2}$
CFS5MM	23.1	$2.05\cdot10^{-4}$	91.2	$3.43\cdot10^{-3}$	-	-	344	$1.44\cdot10^{-2}$
CFS6LM	23.2	$8.34\cdot10^{-5}$	96.9	$2.03\cdot10^{-3}$	236	$3.82\cdot10^{-3}$	343	$1.88\cdot10^{-2}$
CFS6MM	22.7	$2.40\cdot10^{-4}$	90.8	$4.98\cdot10^{-3}$	236	$3.36\cdot10^{-3}$	343	$1.70\cdot10^{-2}$

[1] The pectin peak was identified as the local peak maximum between 200 °C and 286 °C for the CFS3 samples or between 200 °C and 256 °C for the CFS6 samples.

[2] The cellulose peak was identified as the local peak maximum between 300-450 °C.

Table S5. *GAB model parameters for the citrus fibre samples (CFS1-CFS6) at room temperature.*

	M_0 (g/100 g)[1]	C[2]	K[2]	R^2
CFS1	6.14	10.9	0.79	0.9994
CFS2	5.10	23.3	0.83	0.9970
CFS3	5.84	13.6	0.79	0.9997
CFS4	5.07	22.5	0.83	0.9982
CFS5	4.89	19.8	0.81	0.9981
CFS6	5.88	22.2	0.80	0.9962

[1] M_0 is the moisture content in the monolayer (g water/100 g dry solids).

[2] *C* and *K* are temperature-dependent constants.