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# Supporting information:

# Towards natural-fibre-based thermoplastic films produced by conventional papermaking

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## Effect of pressing conditions on optical appearance and density

The optical transparency after pressing was increased by increasing the pressing temperature to about 150 °C (Figure S1a) whereas the density increase levelled off at about 110 °C (Figure S1b). No improvements in optical properties could be detected beyond 150 °C, 16 MPa pressure and a pressing time of 2 min, either with the UV-vis instrument or by the human eye.





Figure S1. (a) light transmittance and (b) density of sheets hot-pressed under different pressures, temperatures and times. Densities exceeding 1500 kg/m<sup>3</sup> presented thinning and raised edges.

## Fourier transform infrared spectroscopy (FTIR)

As support to carbonyl determination after reduction with borohydride, FTIR was used to show that the reduction do dialcohol cellulose was complete. FTIR was performed on a Perkin-Elmer Spectrum 2000 FTIR equipped with an attenuated total reflectance crystal accessory (Golden Gate). Figure S2 clearly shows that no carbonyls were present in the final material, i.e. no peak at 1740 cm<sup>-1</sup>. Furthermore, the FTIR spectra also show that there is a decreased absorbance at the wavenumber around 1110 cm<sup>-1</sup> corresponding to secondary alcohols, i.e. supporting formation of dialcohol cellulose.



Figure S2. FTIR spectra of oxidised-reduced papers in the wavenumber interval of (a) 600–4000 cm<sup>-1</sup> and (b) 800–1800 cm<sup>-1</sup>. The dotted and dashed line in (b) corresponds to wavenumbers commonly assigned to carbonyls and secondary alcohols, respectively.

#### Optical properties at different degrees of modification

Table S1. Optical properties of oxidised-reduced papers, measured at a wavelength of 550 nm, before and after pressing for 2 min at 150 °C and 16 MPa. Full spectra can be found in Figure S3.

	Before pressing			After pressing		
	Average	Total	Haze	Average	Total	Haze
	thickness	transparency	(%)	thickness	transparency	(%)
	(µm)	(%)		(µm)	(%)	
Reference	183 ± 5	18.1 ± 0.5	98.8 ± 0.4	150 ± 2	19.5 ± 0.3	98.8 ± 0.4
6 h beaten	132 ± 1	40.4 ± 1.9	98.1 ± 0.3	113 ± 3	47.1 ± 2.9	92.7 ± 2.5
12 h beaten	124 + 6	77.6 ± 0.4	88.8 ± 0.4	105 ± 6	86.7 ± 1.1	32.3 ± 8.6
24 h beaten	125 ± 3	78.6 ± 1.1	87.0 ± 0.5	108 ± 3	89.4 ± 0.5	22.8 ± 6.5
12 h non-beaten	147 ± 1	34.6 ± 2.1	98.9 ± 0.3	116 ± 3	49.8 ± 3.2	90.8 ± 2.5
24 h non-beaten	117 ± 1	75.6 ± 1.3	88.4 ± 0.6	100 ± 1	87.2 ± 0.4	33.5 ± 8.9



Figure S3. Total transmittance and haze of oxidised-reduced sheets as a function of wavelength for fibres of different degree of oxidation; (a) before pressing and (b) after pressing at 150 °C, 16 MPa for 2 min.

#### T-peel tests of modified sheets hot-pressed together

The adhesion between two hot-pressed papers was evaluated by a t-peel test (Figure S4a). Two 20 mm wide strips of paper made from the most modified fibres were placed on top of each other and fused by hot pressing of a 20 mm by 63 mm area in the middle of the two strips, so that four free ends were left non-fused (the cloudy parts seen in Figure S4b). The fused strips were then cut in the middle of the fused area into two test pieces with two free ends each. Figure S4c shows force-displacement curves for the modified paper and, to sever as a reference, Figure S3d shows force-displacement curves for separating two sticky notes, showing that a 10–15 times greater force is needed to separate the fused papers than peeling of a sticky note.



Figure S4. (a) Schematic setup of t-peel test, (b) photo of test-pieces, (c) peel-force curves separating two hot-pressed pieces of oxidised-reduced paper (150 °C, 16 MPa for 2 min), (d) peel-force curves separating two sticky notes.