

## Supporting Information

*Direct Ink Writing of fully bio-based liquid crystalline Lignin/Hydroxypropyl Cellulose aqueous inks: optimization of formulations and printing parameters*

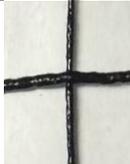
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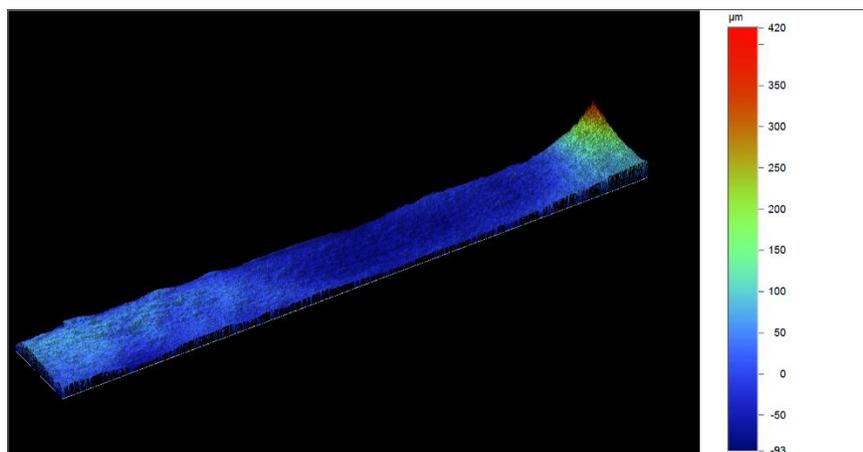
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**Table S1: Pre-screening test of bio-inks with an OSL solution of 60 %**

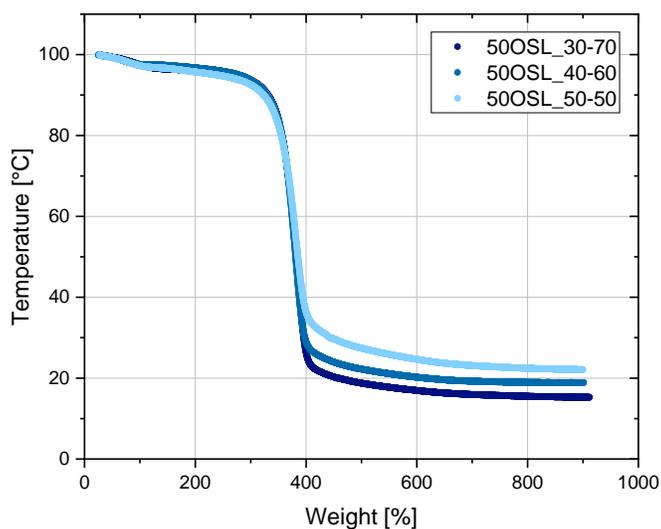
		60 % OSL solution					
OSL:HPC	30/70	35/65	40/60	45/55	50/50	55/45	60/40
Fiber formation							
Layer stacking							



**Figure S1: Surface profile of sample 47.5OSL\_30-70**

**Table S2: Results of F-tests for different parameters and factors**

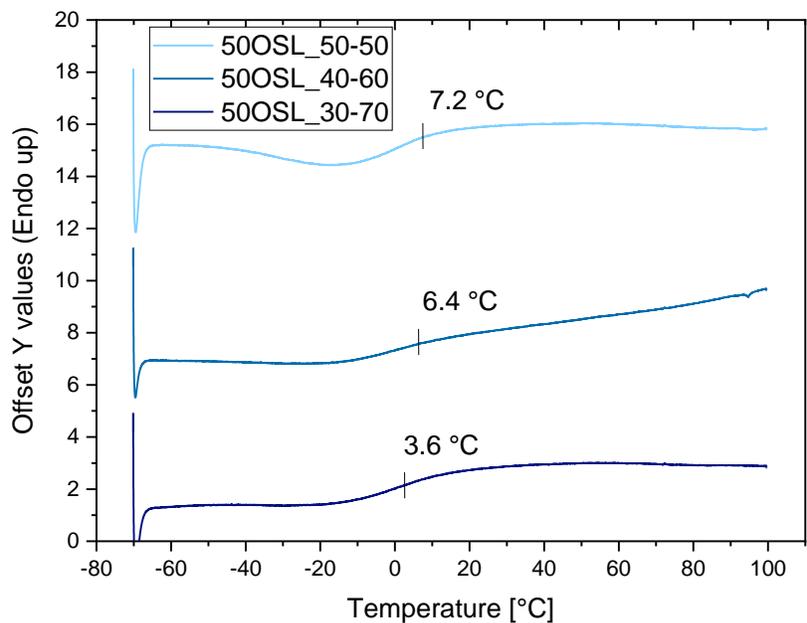
Parameter	Solid Content	OSL/HPC ratio	Printing speed	Pressure
Modulus	0.025	0.003	0.790	0.773
UTS	0.002	0.031	0.485	0.057
Toughness	0.287	0.046	0.346	0.005
Elongation	0.091	1.6e-6	0.668	0.284
Dim. error length	0.001	5.6e-8	0.505	0.195
Dim. error width	0.726	0.009	0.880	0.008
Dim. error thickness	2.2e-16	0.003	0.446	0.375



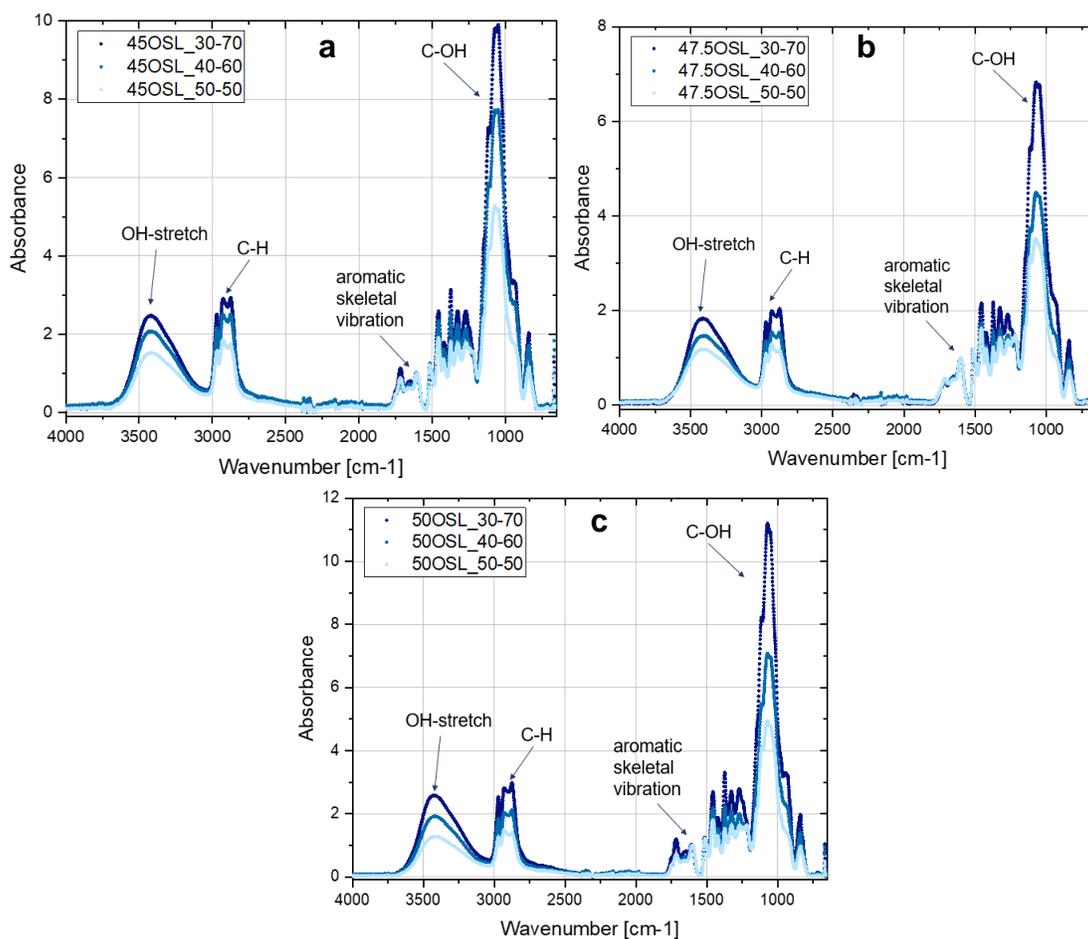
**Figure S2: Typical TGA curves of samples with 50 % lignin solution at different ratios**

**Table S3: TGA results of printed samples**

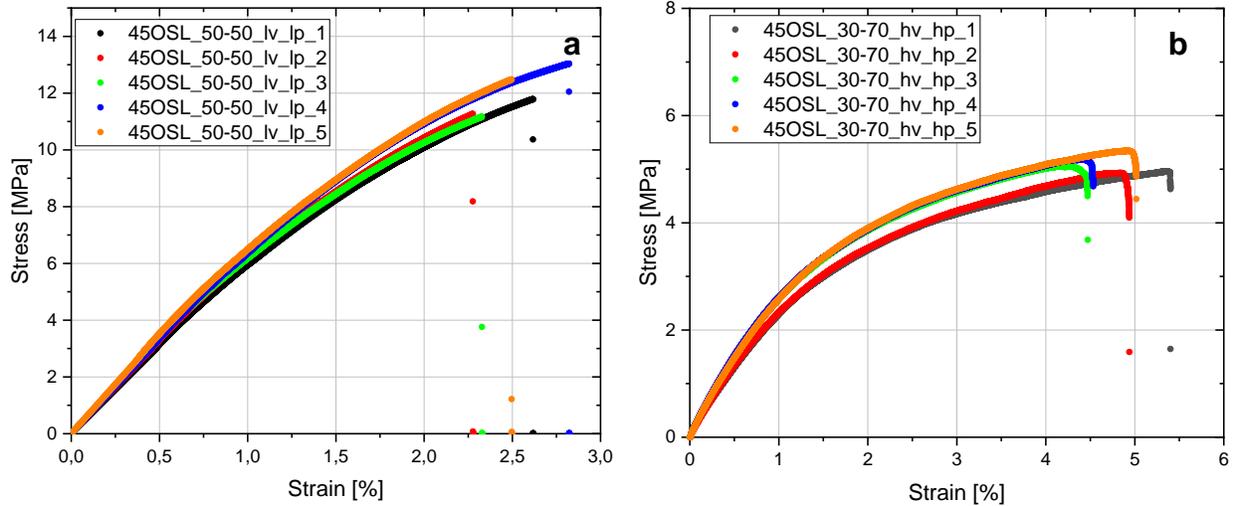
Sample	Water content [%]	T <sub>degradation</sub> [°C]	T <sub>onset</sub> [°C]	Residual mass [%]
45OSL_30-70	3.5	395	371	14.3
45OSL_40-60	2.5	389	371	18.9
45OSL_50-50	3.5	370	360	23.7
47.5OSL_30-70	2.8	394	310	11.0
47.5OSL_40-60	2.8	382	368	21.3
47.5OSL_50-50	3.3	380	364	21.7
50OSL_30-70	3.1	386	355	15.3
50OSL_40-60	2.4	382	358	18.9
50OSL_50-50	3.1	381	366	22.1



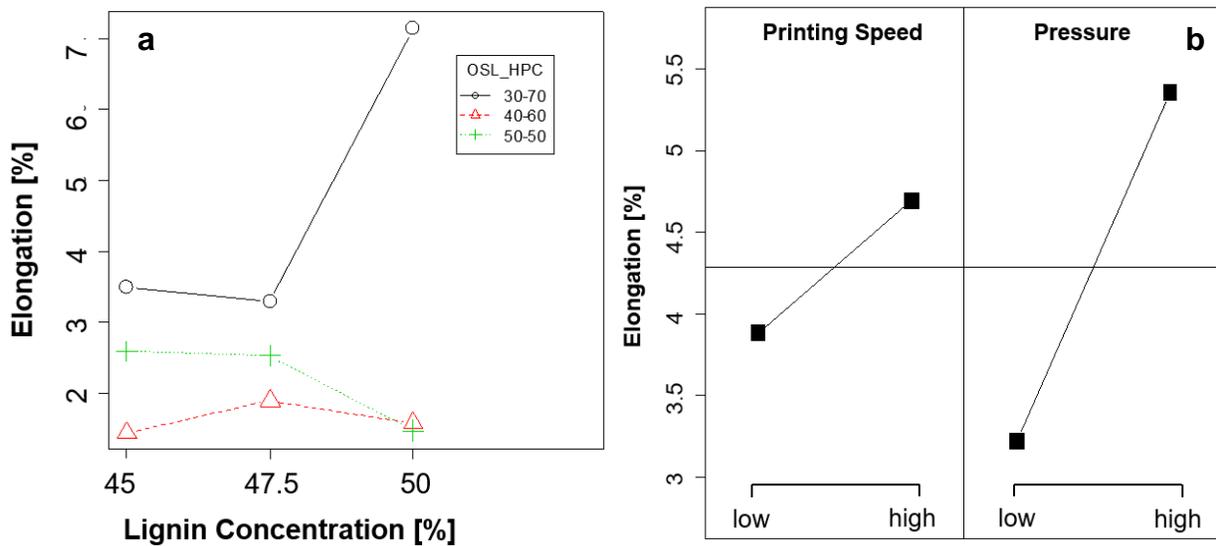
**Figure S3: Effect of blend composition on thermal properties of 3D printed samples (numbers show Tg of respective blend)**



**Figure S4: FTIR spectra of printed samples with lignin solutions of 45 % (a), 47.5 % (b) and 50 % (c)**



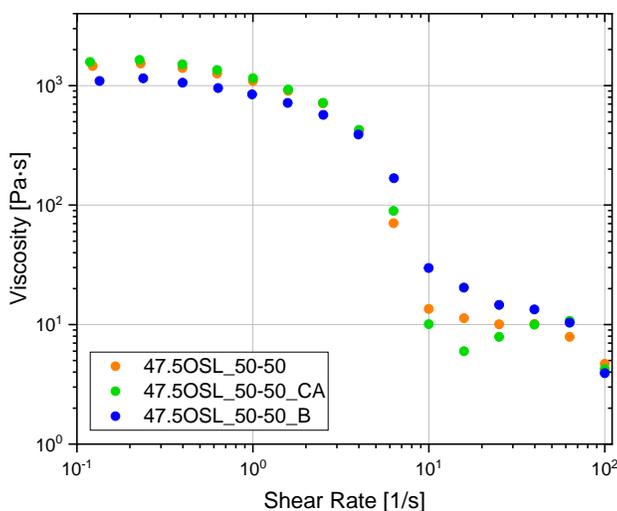
**Figure S5: Representative stress-strain curve for five replicates of sample 45OSL\_50-50\_iv\_lp (a) and 45OSL\_30-70\_hv\_hp (b)**



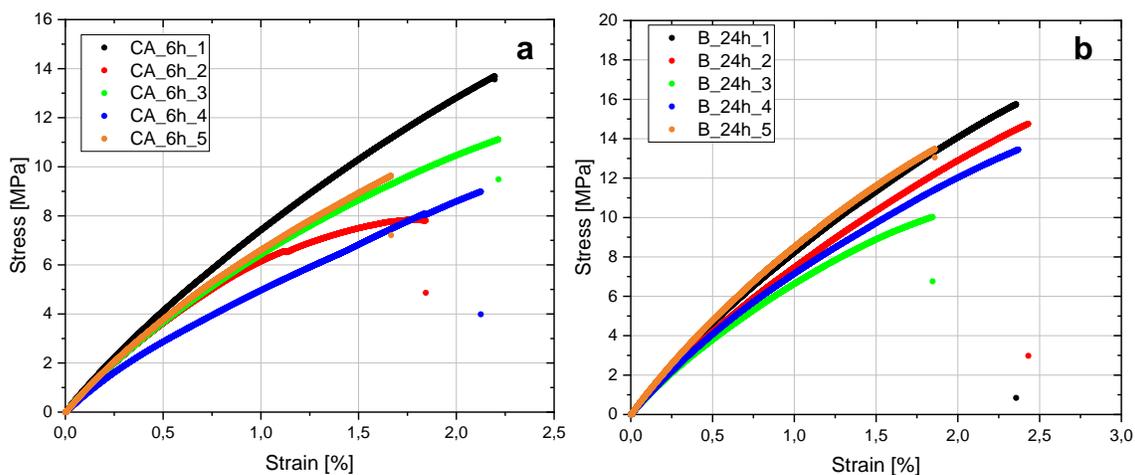
**Figure S6: Main effect plots of elongation – impact of composition (a) and printing parameters (b)**

**Table S4: Swelling test of cross-linked samples in comparison to a sample without cross-linker (47.5OSL\_50-50)**

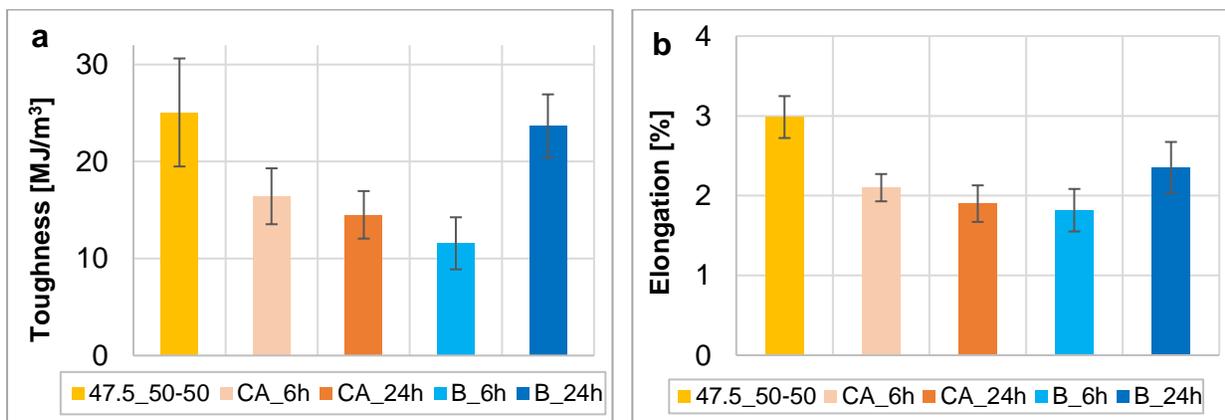
Sample	Gel content [%]
47.5OSL_50-50	64.6
CA_6h	81.9
CA_24h	79.7
B_6h	78.4
B_24h	76.1



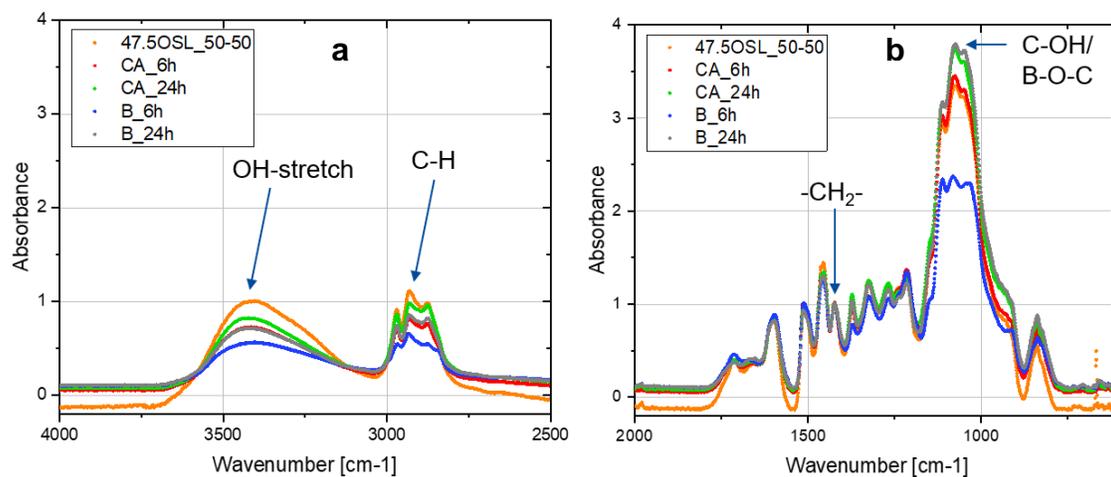
**Figure S7: Flow curve of samples containing citric acid and borate ions as cross-linkers**



**Figure S8: Representative stress-strain curve for five replicates of sample CA\_6h (a) and B\_24h (b)**



**Figure S9: Toughness (a) and elongation (b) of cross-linked samples in comparison to a sample without cross-linker (47.5OSL\_50-50)**



**Figure S10: FTIR spectra of cross-linked OSL/HPC samples for a region of 4000 to 2500 cm<sup>-1</sup> (a) and 2000 to 600 cm<sup>-1</sup> (b)**