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Supporting Information

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Additive Manufacturing of 3D Structures Composed of Wood Materials

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Technologies	Techniques	Fabrication approachUnique properties		
Molding	RT drying	Samples were left to dry at RT	High-density structure	
	Freeze- casting	Mold samples formed on cold controlled stage followed by lyophilization	Aligned foam structure, low- density structure	
Extrusion	DIW	Left to dry at RT, multiple extruders possible	High-density structure	
	DCW	3D print onto a cold, controlled stage, followed by lyophilization	Aligned foam structure, low- density structure, thermal insulator	
Inkjet	Drop on Demand (DoD) Inkjet	2D printing on a substrate	Patterned layer on a substrate	
	Binder-jet	Inkjet printing of CNC/XG onto continuous layer of WF	Low-density structure and thermal insulator	

 Table S1. Different approaches used to fabricate wood-based structures.

Name	CNC [wt.%]	XG [wt.%]	WF [wt.%]	Ref
5% WF	9.1	0.075	5	
11.5% WF	8.5	0.069	11.5	DIW/DCW
14.5% WF	8.2	0.067	14.5	F1g 51-3
30% WF	6.7	0.055	30	
DW	0	0	20	
0:1	1.2	0	20	
1:100	1.19	0.01	20	Fig S9
1:10	1.18	0.02	20	
1:4	1.1	0.1	20	
3%	3	0.3	9	
2%	2	0.2	9	
1%	1	0.1	9	Fig S10
0.5%	0.5	0.05	9	
0.1%	0.1	0.01	9	
Pine CNC-LAB	3.5	0.07	11.5	
Maple CNC-LAB	3.5	0.07	11.5	DIW/DCW
75 CNC-LAB	3.5	0.07	11.5	Fig S11
Pine CNC-CF	8.2	0.07	11.5	DIW/DCW
Maple CNC-CF	8.2	0.07	11.5	Fig S12
Name	CNC [wt.%]	XG [wt.%]	BYK 348 [wt.%]	Ref
Inkjet-CNC	1.5	0	0	Inkjet
Binder-CNC	1.5	0	0.011	Binder jet
Binder-XG	0	0.1	0.01	Fig S13

Table S2. Ink compositions



Figure S1.

Rheology measurements of inks containing different concentration of WF (75, Pine, Maple) while the XG:CNC (1:122) ratio was kept constant. (**Top**) All inks exhibiting shear thinning behavior are presented, as are (**Bottom**) unprintable inks (too liquid or too viscous).



Figure S2.

Oscillation stress sweep of printable inks at f=1Hz. (**Top**) Low phase angles indicate "solid-like" behavior while high phase angles indicate "liquid-like" behavior. (**Bottom**) Storage modulus indicating linear visco-elastic region up to ~100 Pa.

% WF	WF source	Yield stress point [Pa]
11.5%	75	904
	Pine	200
	Maple	263
14.5%	75	414
	Pine	309
	Maple	336

Table S3. Yield stress point (flow point) derived from oscillation stress sweep.



Figure S3.

Oscillation frequency sweep of printable inks at τ =10Pa.



Figure S4.

Cross-section images of engineered woods and a 3D-printed wood-based sample (DIW).



Figure S5.

Close-up photo of Fig 1C, 3D DIW printing of multi-material. (A) Image showing final printed structure and (B) microscopy image showing smooth bonding between the two types of wood.



Figure S6.

Microscopy images of CNC-CF ink jet drops and patterns printed on a silicon wafer: (**A**,**B**) 200 dpi, (**C**) 1 layer at 600 dpi and (**D**) 5 layers at 600 dpi.



Figure S7.

Optical profile measurements of CNC-CF ink jet drops on a silicon wafer substrate at (**Top**) 200 dpi, showing uniformity reproducibility of droplets. (**Bottom**) Morphology measurements of a single drop: diameter of 55 μ m and height of 40um at the center of the droplet and height of coffee ring effect edge was 75 nm.



Figure S8.

(A) AFM image of a single CNC-CF ink jet drop on a silicon wafer substrate (B-C) zoom-in measurements.



Figure S9.

(A) Stress-strain curves and (B-C) modulus and compressive strength derived from compression testing of Eucalyptus mold samples at different XG:CNC ratios (Table S2). (D) Photograph of molded wood samples.



Figure S10.

(A) Photograph of freeze-cast molded wood foam samples. (B) Stress-strain curves and (C-D) modulus and compressive strength of samples prepared from inks with different binder:WF ratios, all containing a 1:10 XG:CNC ratio (Table S2).



Figure S11.

(A) Photograph of printed samples (left: '75'; right: 'maple') (B) Stress-strain curves and (C-D) modulus and compressive strength derived from compression tests of wood DIW-printed with ink comprised of CNC-LAB and different concentrations of WF (Table S2).



Figure S12.

(A) Photograph of DIW printed samples (B) Stress-strain curves and (C-D) modulus and compressive strength derived from compression tests performed on wood DIW-printed with ink comprised of CNC-CF and different concentrations of WF (Table S2).



Figure S13.

Dependence of surface tension on surfactant concentration in Binder-CNC and Binder-XG inks (Table S2).