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ORIGINAL PAPER

Wood-based 3D printing: potential and limitation to 3D print building elements with cellulose & lignin

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Abstract

Under urgent sustainability targets, the building industry craves for renewable and recyclable biomaterials as cellulose is a fiber; Lignin is a plant-derived low-cost polymer with remarkable properties, yet its valorization is in its infancy. Recent studies have shown potentials to combine cellulose and lignin into a renewable bio-based material for the built environment, with the use of additive manufacturing to allow geometric customization and local control of material. However, previous studies also highlighted crucial issues to be solved. One main challenge is the lack of knowledge on combinations of lignin and cellulose with different binders to achieve a paste suitable for 3D printing, leading to a material applicable in the built environment. To contribute overcoming the challenge, this research aimed to explore various combinations of cellulose, lignin, and binders and to study the extrudability of the resulting paste using a clay extruder installed on a robotic arm. Several combinations were explored, evaluated, and compared. The four recipes with the highest scores were used to produce samples for tensile and three-point bending tests, water absorption and retention tests, and microscope analysis. The overall outcome has shown similarities between the mechanical properties of the mixture developed using methylcellulose as the binding agent and rigid polymer foams, such as the ones commonly used as insulation panels. Moreover, the material mix with the highest score in the preliminary assessment was further applied to fabricate samples with varied geometries to assess its potential and limitations combined with the fabrication process. Finally, two demonstrators were produced to explore the printing process for different geometric configurations: conceptual window frame and structural node were designed, and 3D printed as proof of concept.

Keywords Wood · Lignin · Cellulose · 3D printing · Window frame · Structural design

Introduction

The built environment is a major contributor to the greenhouse gas emissions, reaching 40% of the global amount [1]. The rising population, climate change and increasing depletion of natural resources create a panorama where the construction industry has the responsibility of acting quickly to this potentially catastrophic scenario [2]. This implies reconsidering the construction supplies; for which natural materials such as hardwood, softwood and bamboo are estimated to be crucial in the following century [2]. These are renewable resources, but with limited environmental impact only if well managed and sourced from reforestation areas [3]. Still, every year more than 10,000 km2 of land, partially from deforestation areas, are converted into fast-wood plantations to feed the timber industry [4], increasing the environmental impact, and reducing the biodiversity [5]. Approximately 37,500 km2 [6] of forests were lost only in 2021 – as a comparison, the area of the Netherlands is 41,850 km2 [7]. Meanwhile in Europe, 25ton of wood are discarded annually [8]. Such residues, combined with agricultural and paper waste, present difficulties to be directly repurposed without processing, however they are rich in the two main building blocks of timber – cellulose and lignin [9].

Lignin and cellulose are the most abundant organic polymers on earth [9] and present a renewable alternative

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to petrochemical-based products [10]. Biorefineries produce more than 50 million tons of lignin worldwide every year, of which 98% are used for energy generation. Only 2% of the total amount are valorized and repurposed in the industry, mainly in the composition of dispersants, adhesives, and fillers [10]. A substantial manner to exploit the potential of waste materials into an object is the use of an additive manufacturing process. Wood as a feedstock has been investigated since at least ten years [11], including its building blocks cellulose and lignin, usually combined separately with bioplastics and other additives. Experiments on the use of both polymers combined are recent. Currently, they are subject of the Wood Without Trees research line at TU Delft, initiated with a graduation project [12] that introduced the topic and identified the material potential in the architecture field. The study presented in this paper is part of this research line and focused on the further development of a feedstock, defining an adequate binding agent, understanding its properties, and applying it to a prototype fabrication. On a larger scale, the aim is to envision an architecture that is customizable at a low cost, while being sustainable and fabricated efficiently.

Background

Cellulose & Lignin

Wood consists of three basic elements – carbon, hydrogen, and oxygen [12] – which form its main three components – cellulose, hemicellulose, and lignin [13].

Cellulose is the most abundant polymer in nature, encompassing between 33 and 51% of the total composition of wood [14]. It is a structural component of the walls in any plant [15], with great tensile strength and hygroscopic behavior [16]. Cellulose has been used centuries ago in many fields, from the generation of energy to clothing and paper production, summing up to 1.5 trillion tons of biomass annually [16]. The fibers for pulp production and paper industry are mainly sourced from wood and plants through the kraft processing of wood logs [16], commonly associated to large environmental impact. Alternatively, they can be obtained from wood waste, such as MDF and HDF boards [17] and paper waste [18] through chemical modification, retaining similar properties to the fibers obtained through the previous method. Agricultural waste, such as pineapple, banana, and sisal biomass, is also a vast source of cellulose [19], together with sugarcane bagasse [20] and rice husk [21].

The second most abundant polymer in nature is lignin, accounting for a share between 21 and 32% of the total composition of wood [14]. It is responsible for the physical strength of the walls of any plant, although alone it is brittle and presents a hydrophobic behavior [22]. Similar to the cellulose fibers, its molecules start to break at 100 °C, quickly degrading above 225 °C [23]. Lignin is largely sourced as a by-product from cellulose extraction, obtained from the residual black liquor from the wood pulping process [24], summing up to 100 million dry tons annually [25]. It is also derived from biorefinery processes of agricultural and wood waste [26], reaching a volume of 50 million dry-tons per year [10]. From this amount, 98% of the biomass is burnt to produce energy. The remaining 2% are used to produce dispersants and adhesives.

Wood-based additive manufacturing

Sawdust, combined with binding agents, has been experimented as a feedstock in additive manufacturing with limited bio-based content, mixed with either synthetic resins [11] or HDPE [27].

In the research developed by Henke & Treml [28], wood powder was combined with gypsum, methylcellulose, sodium silicate and cement, and used to fabricate a truncated cone by depositing layers of thin particles and spraying water to activate the binders. In another study, it was mixed with commercial adhesives produced with polyvinyl acetate (PVAc) and urea–formaldehyde (UF) and used to 3D print samples with a wood content of 25% [29]. In the study developed by Rosenthal et. al. [30], this percentage increased to 89% with the formulation of a gel-like solution with methylcellulose, applied to the production of simple cylindrical geometries. The ground-braking mark was the research developed at Umea School of Architecture [31] which resulted in a material with 85% of wood content, mixed with methylcellulose and bentonite, and used in the fabrication of a 60 × 45 cm vase.

Combined with polymer matrices, such as polylactic acid (PLA), wood powder has also been used to produce filaments for liquid deposition (LDM) processes [32], with content ranging from 5 wt.% [33] to 50 wt.% [34]. However, it was observed that a concentration of 20 wt.% or less maximized the mechanical properties of the material as well as the extrusion smoothness [35].

Cellulose-based additive manufacturing

Cellulose has already been introduced as a feedstock for aerogels and hydrogels in the bio-printing and tissue engineering fields [13]. Generally, the main interest around the polymer regards its mechanical properties and high potential as a natural fiber reinforcement in polymer matrices. An example of its use is the fungal-like additive material – FLAM [36]. Based on the walls of the oomycetes, it combines chitin with cellulose creating an extrudable paste with similar mechanical properties to low-density woods [36]. Combined with resin, cellulose was also used to develop

a novel material and fabricate a 2 m-tall structural wall at Chalmers University [37].

Lignin-based additive manufacturing

Lignin has already been used to produce inks and filaments for FDM fabrication, mixed with thermoplastics in ratios between 40 and 70% [38] and used to prototype small objects, such as phone cases [10]. These filaments are often difficult to manipulate and too brittle, although when chemically modified and integrated with either acrylonitrile butadiene styrene (ABS) or nylon12, lignin creates a strong composition with excellent printability [25]. Both matrices have been investigated by Nguyen et al. [23], blending them with the polymer at ratios of 40 wt.% and 60 wt.% with and without carbon fiber reinforcements. Favorable for the printing process, the results have shown improvements in its stiffness and melting viscosity at room temperature. The increase in the tensile strength validated the polymer's reinforcement potential.

Cellulose & lignin-based additive manufacturing

Cellulose and lignin have been exploited as reinforcement fibers and fillers, respectively and separately [15]. Combining both polymers in the same mixture is a relatively new field, and the state-of-art research was published by Thomas Liebrand in "3D printed fiber reinforced lignin" [12]. The experiments were based on the mixture of kraft lignin and kraft cellulose sheets, bleached, which were pulped and combined with demineralized water and acetone at different proportions to create an extrudable material. The study validated the material potential as a novel wood-based material for architectural applications, [12] however the toxicity and flammability of acetone [39] implies that any further developments on the material would require exploring alternative binding agents.

Binders

The exploration of cellulose in combination with lignin demands a binding agent to create a reinforced matrix. Adhesives in general, based on PVAc and UF, have been successfully used as binders, mixed with wood-based particles and powder, and applied in additive manufacturing [29]. As a natural alternative derived from the food industry waste, Bone glue is conventionally used either as an adhesive in woodwork or as an additive in biopolymer concrete mixtures [40].

Known applications of cellulose in the food industry [41] and the ancient use of starch as an adhesive [42], raised the possibility of investigating the food spectrum for promising alternatives. Baking soda modifies the pH of water, creating an alkaline solution which favors the dissolution of lignin [43], Vegetable oils were identified as additives to enhance the mechanical properties of bio-based composites when combined with natural fibers [44]. Xanthan gum, a natural thickening agent [45] has been used to create multiple edible gels for additive manufacturing, mixed with starch [46] and cellulose powder [47]. Starch is used in bio-adhesives as a harmless alternative to toxic formaldehydes [48], presenting as a great candidate for green adhesives coming from the wood industry [49].

The numerous applications of cellulose in the biomedical industry [13], also suggested a few alternative binders. Dimethyl Sulfoxide (DMSO) is used as a solvent for cellulose fibers in the fabrication of hydrogels for additive manufacturing [50] Glycerin is used to control the viscosity of cellulose-based gels for biomedical applications and optimize its extrusion [51].

Commonly found in marine brown algae and soil bacteria, alginate is used as a binding agent for fillers, including sawdust [52], forming a viscous solution when mixed with water [53]. Beeswax is also used to produce bio-based solutions, mixed with cellulose and lignin to create protective coatings for paper [54].

Methylcellulose has been traditionally used as an additive to produce cement, adhesives, and mortars, to increase the viscosity and improve the homogeneity, cohesion, and workability of the recipe [55]. In the additive manufacturing industry, it has been used as an additive to enhance the viscosity of cellulose-based hydrogels [56] and to create homogeneous and extrudable materials based on wood powder [30] and cellulose and bentonite [31].

Research methodology

The study presented in this paper was divided into five main phases – background, material exploration, material properties exploration, printability exploration, and design & prototyping – combining a theoretical and practical framework.

The background research followed a traditional approach through a mix of offline and online searches in libraries and search engines, forming a panorama of the material and fabrication methods within the industry. The state of the art was defined, and novel binders and additives were identified.

As the exploratory steps commenced, material recipes were developed and documented. Mixes combining the main raw materials – cellulose and lignin – with the selected binders and additives were developed and tested at eyesight and touch and evaluated based on nine parameters – homogeneity, viscosity, adhesion, extrudability, bio-based content, shrinkage, brittleness, curing time, aesthetics. A value of -1, 0 or 1 was given to each mixture for a negative, indifferent, or positive performance, respectively. The final grade

for each recipe offered a comprehensive comparison among the produced samples, determining the four most promising recipes to be further investigated.

The material properties exploration phase followed with the aim to benchmark the discovered recipes with wood-based materials in terms of their mechanical properties for additive manufacturing. Test specimens were made through a molding and extrusion process and later tested. Four different tests were executed: water absorption and shrinkage, flexural modulus and strength, tensile modulus and strength, and microscope analysis.

In parallel, the printability exploration proceeded by attempting to fabricate samples with common and simple designs to assess the material and equipment limitations, using the material that had the highest potential. The work was executed at the Laboratory for Additive Manufacturing in Architecture – LAMA – at the Faculty of Architecture and the Built Environment of TU Delft, using the available clay extruder and robotic arm.

Concluding the study, the design & prototyping phase focused on the fabrication of relevant components for the construction industry. Last refinements were executed on the mixture and printing process, derived from the outcome of the exploratory phases. A structural node and a section of a window frame were selected to explore the potential and challenges previously identified (Fig. 1).

Material parameters

The material exploration commenced by analyzing cellulose and lignin individually and combined, assessing their melting and degradation points. It continued with the reproduction of the state-of-the-art mix containing cellulose, lignin, and acetone [12] and defining a benchmark for the experiments. At last, the solvent was replaced with the binding agents researched—xanthan gum, methylcellulose, DMSO, corn starch, alginate, glycerin, bone glue, wood glue, vegetable oil, baking soda, beeswax. Samples were produced and a comparison chart was elaborated, based on the established parameters and evaluation (Fig. 2).

The experiments were executed at three locations – LAMA, StevinLab and the Model Hall of the Faculty of Architecture and the Built Environment of TU Delft – with similar environment conditions, measured with a DHT11 digital sensor linked to an Arduino kit. Room temperature was kept between 20˚C and 23˚C and relative humidity between 37 and 45%.

The material mixes were prepared by blending the cellulose at first to separate the fibers and reduce clots. Lignin was added in the sequence and continuously blended for 15 min to evenly cover all fibers with the polymer. Water and binder were the last ingredients to be incorporated, mixing

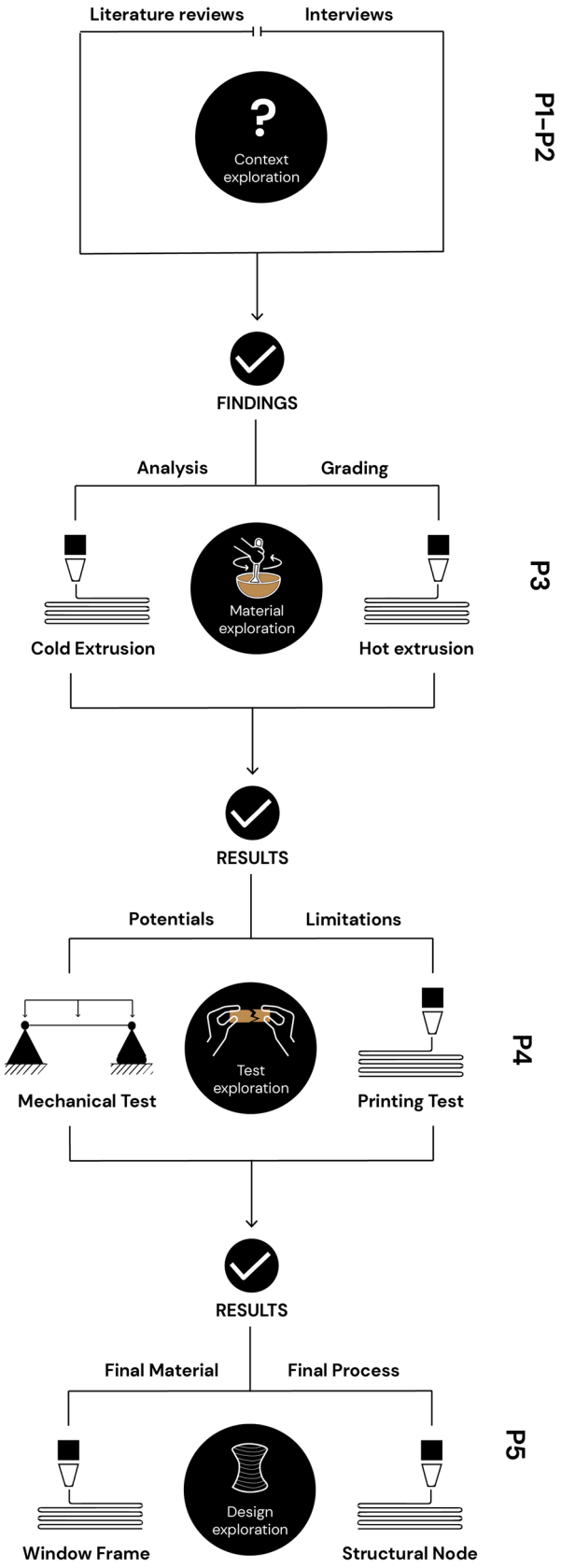

Fig. 1 Workflow Overview (diagrams by authors)

Fig. 2 Material Experimentations in a hot and cold extrusion process (Pictures by authors)

until homogeneous. The process required an enclosed receptacle to reduce material losses due to the chalky aspect of the lignocellulosic polymers. Despite difficult to quantify, a compensation of 5% on the ingredients quantities was adopted.

Cellulose and lignin rapidly deteriorated when heated above 130˚C, charring instead of melting. When mixed with water, lignin created a brittle material meanwhile cellulose absorbed the liquid and clustered. Both polymers combined with water resulted on a low viscosity matrix and poor bonding with the fiber reinforcement, resulting in a thick liquid filled with fiber clots. This outcome demonstrated that a binding agent is necessary to combine both polymers into a viscous substance.

Lignin was successfully dissolved with acetone, resulting in a paste with low to medium viscosity and high adherence. The addition of cellulose to the material improved both properties and resulted in long chains of microfibers, meanwhile maintaining the homogeneity. After curing, the samples extruded with a syringe presented outstanding interlayer bonding, however the surface had a rough and porous appearance. The hazardousness of the solvent and its reaction with plastic are the strongest disadvantages of this recipe, demanding a controlled working environment, and glass and aluminum tools for handling and production. The extended hardening time is also a negative aspect, demanding one week to completely cure the samples.

Wood glue combined with cellulose and lignin resulted in a material with outstanding properties in terms of homogeneity, viscosity, and adherence. The reduced amount of water in the recipe reduced the shrinkage and deformation. The curing time observed was short for the surface – 24 h – although the samples demanded one week to harden entirely. The disadvantage of this mixture was the PVAc-based adhesive, which hinders a full bio-based formulation.

Most of the natural binders researched and applied in the material exploration did not present satisfactory results when combined and formulated as feedstocks for 3D printing. They did not achieve the adequate viscosity and adhesion for a liquid deposition additive manufacturing process. Xanthan gum, glycerin, and alginate, which create gel-like matrices when activated with water, retained the consistency when mixed with lignin and produced low viscosity substances. Combined with cellulose, they resulted in non-homogeneous materials and failed at reaching minimum viscosity and bonding properties to build multi-layered structures. Methylcellulose was an exception and resulted in the material mix with the best properties among all binders explored. Mixed with water and lignin at temperatures above 80˚C, it formed a homogeneous paste, reaching moderate adhesion and viscosity after cooled down to temperatures below 40˚C. The samples extruded with a syringe presented strong interlayer bonding and a smooth surface appearance resembling wood.

DMSO was the other exception. Mixed with lignin, it created a paste with moderate viscosity and high homogeneity and adhesion, similar to the one resulting from the mixture with acetone. Adding cellulose fibres to the matrix resulted in outstanding adhesion while retaining the homogeneity. However, the samples did not reach a solid state, and their surfaces required an extended curing period, reaching more than four weeks.

A comparison among all the material experiments performed is presented in a summary table (Fig. 3), highlighting acetone, DMSO, methylcellulose and wood glue as the most promising binding agents among all the ones explored. The binder explorations showed that methylcellulose had the less disadvantages, and resulted in a material mix with a full

MATERIAL EXPLORATION SUMMARY & COMPARISON

	HOMOGENEITY	VISCOSITY	ADHESION	EXTRUDABILITY	BIO-BASED	SHRINKAGE	BRITTLENESS	CURING TIME	AESTHETICS	TOTAL
MIX 1 | ACETONE	1	1	1	1	-1	0	1	1	-1	2
MIX 2 | XANTHAN	-1	-1	-1	-1	1	0	0	-1	-1	-5
MIX 3 | MC	1	1	1	1	1	-1	0	1	1	6
MIX 4 | DMSO	1	1	1	1	1	0	0	-1	0	4
MIX 5 | GLYCERINE	-1	-1	-1	-1	1	0	0	-1	0	-4
MIX 6 | CORN STARCH	0	-1	-1	-1	1	0	-1	0	1	-2
MIX 7 | ALGINATE	-1	-1	-1	-1	1	0	0	0	0	-3
MIX 8 | BEE WAX	-1	-1	-1	-1	0	0	-1	1	-1	-5
MIX 9 | BONE GLUE	1	1	1	0	1	-1	0	0	-1	2
MIX 10 | WOOD GLUE	1	1	1	1	-1	0	1	0	1	5
MIX 11 | OIL	1	-1	-1	1	-1	0	0	-1	1	-1
MIX 12 | BS	-1	0	-1	-1	0	0	1	1	0	-1

Fig. 3 Material comparison (diagram by authors)

bio-based composition and the best properties for a liquid deposition additive manufacturing process. Viscosity and adhesion are comparable to the ones from clay, commonly used in such processes, combined with excellent extrudability and homogeneity.

Material properties parameters

The mechanical properties of the four mixes identified as the most promising recipes were assessed and documented to determine their potential applications in the built environment and to compare with other wood-based materials, defining a benchmark in the sector. The ISO standards for polymer testing – NEN-EN-ISO-178 [57] and NEN-EN-ISO-527 [58] – were used as base for the test design, parameters and results interpretation, adapted to the limitations of the novel materials and fabrication process. Currently there are no official standards for the mechanical testing of novel bio-based materials applied to additive manufacturing processes.

Tensile and three-point bending tests were performed to establish the yield stress, the ultimate tensile stress, and the modulus of elasticity at both conditions for all four material recipes. The wood glue-based mix was the strongest one and had the highest values for yield and flexural strength, explained by the material composition based on a synthetic adhesive [29]. The methylcellulose-based mix and the wood glue-based mix were both the stiffest ones, with comparable values for modulus of elasticity. A stronger adhesive based on formaldehydes would potentially improve the mechanical performance of the wood glue-based material [29], however it would incorporate a hazardous component into the formulation.

The acetone-based mix had a poor performance and the lowest values among the samples tested. The highest value reached by any of its samples was lower than the minimum value reached by any other sample from the wood glue or methylcellulose-based mixes. The broken specimens showed a non-homogeneous cross section and a poor matrix coverage on the cellulose fibers. Combined with the brittle, dry and crumbly aspect, these explained the poor mechanical performance of the material mix. The DMSO material samples did not harden entirely, partially retaining a hard gel-like consistency and failing before the test could be executed.

The methylcellulose-based mix reached a maximum modulus of elasticity of 1.05 GPa in bending, and a maximum yield strength of 4.06 MPa. The wood glue-based samples reached a maximum of 0.90 GPa and 4.81 MPa, respectively. For comparison purposes, the modulus of elasticity of elastomers reaches a maximum of 0.1 GPa, meanwhile rigid polymer foams, polymers and natural materials reach between 0.1 GPa and 1 GPa. Ceramics, metals, and composites usually start from 10 GPa. The yield strength of elastomers, rigid polymer foams, polymers, natural materials, and ceramics normally spans between 1 and 10 MPa. Carbon fiber reinforced composites and steel normally have values above 100 MPa, reaching close to 1000 MPa [59]. Therefore, the novel materials developed in this research have comparable properties to the ones of rigid polymer foams.

The effect of water and the requirement of a repellent layer on these novel materials were assessed due to the natural hydrophilic characteristic from both wood and cellulose. The

ISO 62 [60] standards, commonly used for plastics, was used as the base, and adjusted to the material limitations (Fig. 4).

Water absorption and retention tests were executed and verified that similarly to wood, a protective layer is required by all four material mixes to avoid degradation. From the experimented coatings – linseed oil and beeswax – the first one had less impact on the aesthetics of the material, delivering a uniform thin coverage, meanwhile the second one resulted on a thick, uneven, and light-yellow opaque layer, highlighting all surface imperfections (Fig. 5).

The methylcellulose mix was the less dense material observed and performed poorly in terms of water absorption and degradation. The wood glue mix was the denser material observed and presented the best scores among all samples, with smoother surfaces and lower porosity. The acetone mix showed average results and low water retention, unlikely the methylcellulose mix. Overall, a great difference was observed between the samples of uncoated material and the samples with a protective layer, confirming the need of a water-resistant coating.

A final microscope analysis was performed to analyze the homogeneity, porosity, and the fiber behavior – coating, length, and direction. The methylcellulose mix showed the most homogeneous surface and cross section, with a uniform matrix and an even distribution of fibers, all thoroughly coated with lignin. The wood glue mix presented reduced porosity and less homogeneity, with fibers unevenly coated with lignin. The acetone mix had the least homogeneous appearance among all samples; however, it had the longest fibers. The DMSO mix also had a homogeneous cross section, with a rough surface and irregular fiber distribution (Fig. 6).

Fig. 5 Water absorption & retaining test (picture by authors)

Printing parameters

Before attempting any printable tests, the discovered recipes were extruded with a syringe and with an electric caulking gun. Once the extrusion phase was completed with the limitations and potentials of each mixes defined, the subsequent

Fig. 4 Tensile & 3 point bending test (picture by authors)

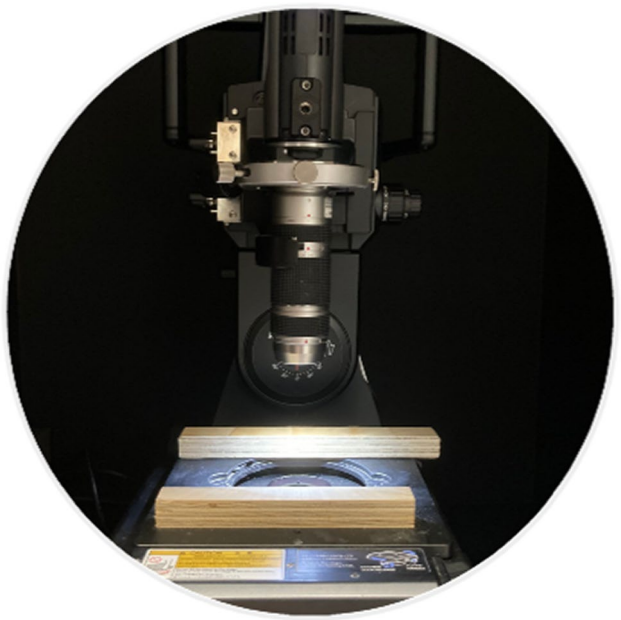

Fig. 6 Microscope analysis (picture by authors)

MECHANICAL PROPERTIES COMPARISON					
	FLEXURAL STRENGHT	MODULUS ELASTICITY (BENDING)	YIELD STRENGHT	MODULUS ELASTICITY (TENSION)	REFERENCE
BEECH, AMERICAN	-	9.5 GPa	86.2 MPa	-	(USDA Forest Service, 2010)
OAK, OVERCUP	-	9.8 GPa	77.9 MPa	-	(USDA Forest Service, 2010)
PINE, EASTERN WHITE	-	8.5 GPa	73.1 MPa	-	(USDA Forest Service, 2010)
SPRUCE, ENGELMANN	-	8.9 GPa	84.8 MPa	-	(USDA Forest Service, 2010)
PARTICLEBOARD	-	2.8 - 4.1 GPa	15 - 24 MPa	-	(USDA Forest Service, 2010)
MDF	-	3.6 GPa	36 MPa	-	(USDA Forest Service, 2010)
OSB	-	4.4 - 6.3 GPa	22 - 35MPa	-	(USDA Forest Service, 2010)
PLYWOOD	-	7 - 8.6 GPa	34 - 43 MPa	-	(USDA Forest Service, 2010)
GLULAM	-	9 - 14.5 GPa	29 - 63 MPa	-	(USDA Forest Service, 2010)
PLA + WOOD POWDER	-	3 GPa	30 MPa	-	(Gardner et al., 2019)
PLA + LIGNIN (40WT%)	-	1.93 GPa	29.25 MPa	-	(Tanase-Opedal et al., 2019)
WOOD POWDER + GLUE	-	3 - 3.94 GPa	30 - 57 MPa	-	(Das et al., 2021a)
TECNARO ARBOBLEND	-	4.3 GPa	58 MPa	-	(www.albis.com)
FLAM!	-	0.26 GPa	6.12 MPa	-	(Sanandiya et al., 2018)
METHYLCELLULOSE MIX	8.59 - 10.60 MPa	0.67 - 1.05 GPa	3.21 - 4.06 MPa	0.33 - 0.56 GPa	(own work)
WOOD GLUE MIX	20.67 - 28.89 MPa	0.64 - 0.90 GPa	3.58 - 4.81 MPa	0.30 - 0.77 GPa	(own work)
ACETONE MIX	4.77 - 9.74 MPa	0.15 - 0.37 GPa	0.81 - 1.51 MPa	0.11 - 0.20 GPa	(own work)

Fig. 7 Mechanical properties overview & comparison (diagram by authors) [21, 61, 10, 32, 62, 36]

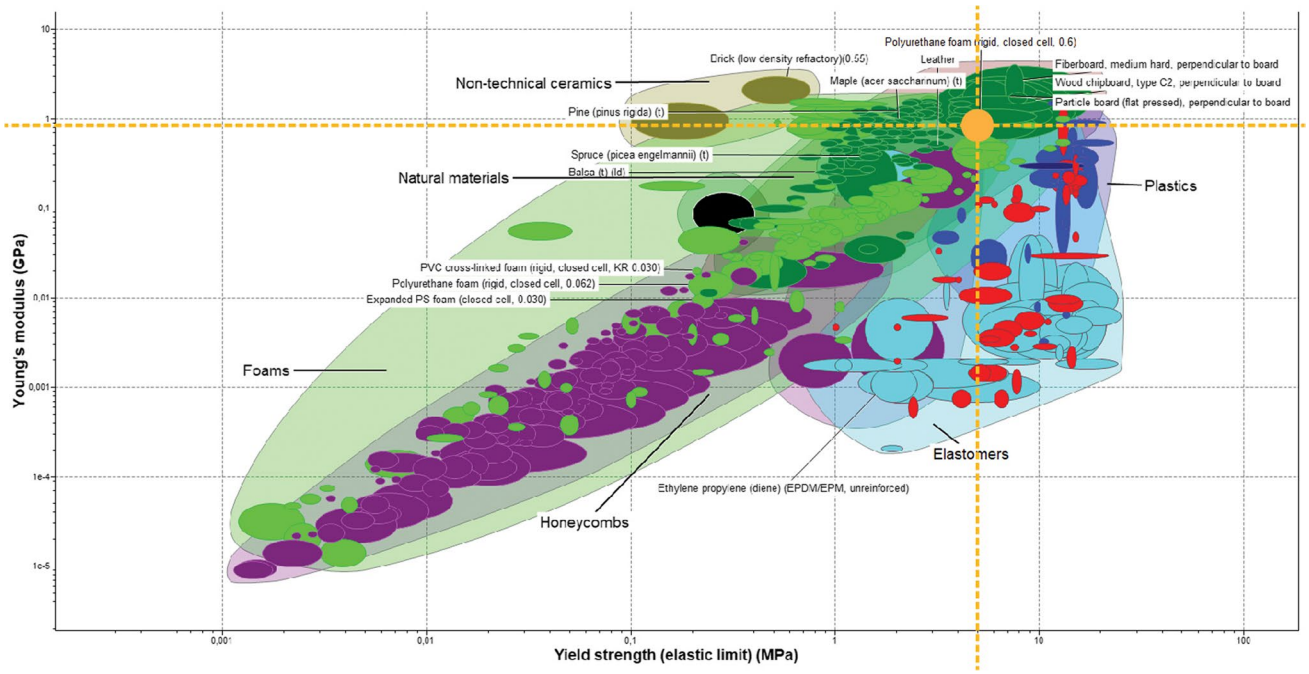

Fig. 8 Methylcellulose based mix & construction materials comparison, created with Granta EduPack 2021 software [59]

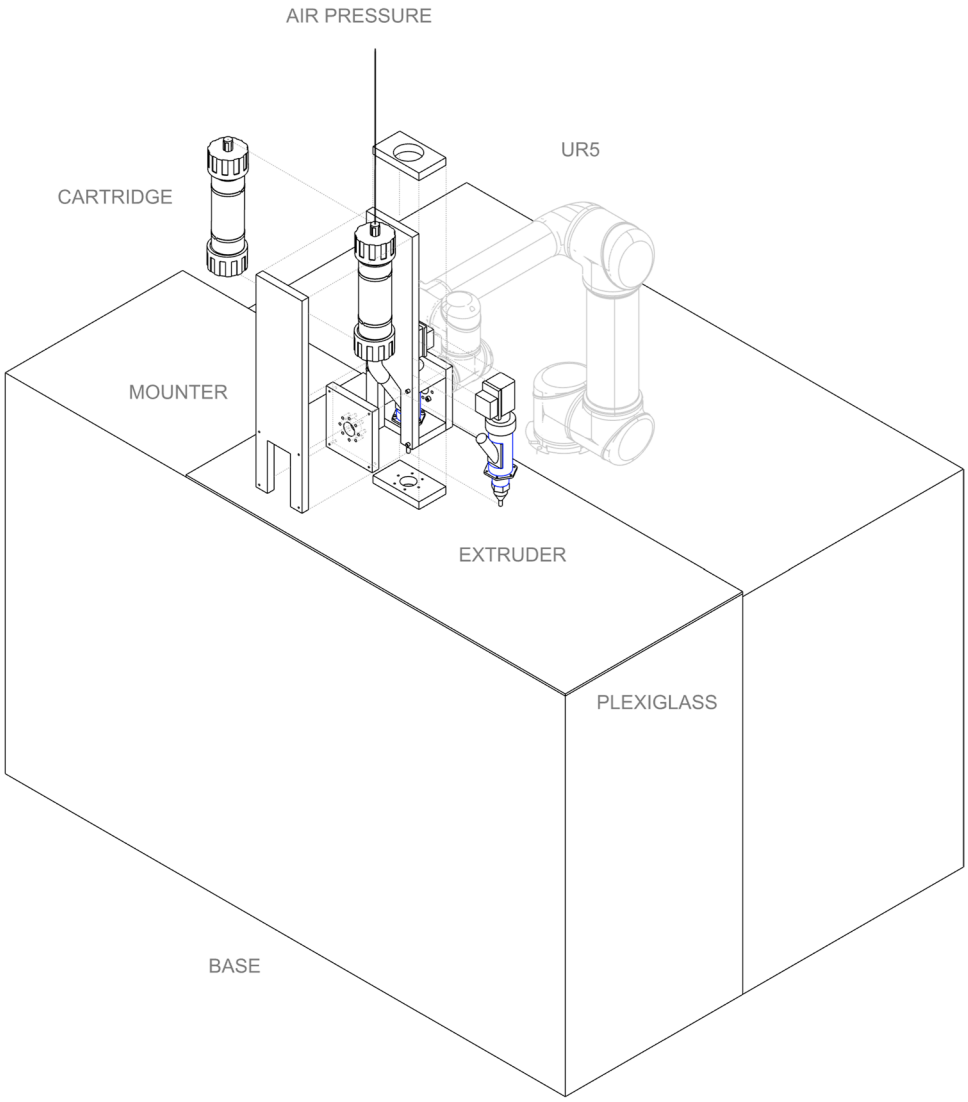

Fig. 9 Printing setup with customized tool (diagram by authors)

step was to explore the additive manufacturing fabrication process of the four most promising mix among the short-listed ones. From this selection, the methylcellulose mix had the highest score as acetone is flammable and hazardous, DMSO requires a curing time to be under 18.5^0C to solidify, and wood glue is chemically based. Thus, methylcellulose was the material of choice for the printability testing. It was a full bio-based and homogenous material which appeared like wood, exerted great viscosity and adherence levels. These elements are fundamental for a liquid deposit modelling fabrication process (Figs. 7 and 8).

The additive manufacturing exploration was performed at LAMA (Laboratory for Additive Manufacturing in Architecture), at the Architectural Engineering and Technology Department of the Faculty of TU Delft. A clay extruder from WASP was made available for the research as well as a 6-axis robotic arm, UR5 (Fig. 9). During the installation of the extruder, the wiring setup and holder of the extruder were custom built onto the robotic arm to further operate the printing tests.

Before achieving any printable tests, the material was mixed until attaining a paste that was flatten onto a vinyl film, which was then pressed and rotated into a cartridge to avoid air bubbles to enter the cartridge and further damage the print. Thus, the weight of the filled cartridge as well as the payload of the robotic arm was measured to obtain the printing length of the model. The position of the nozzle according to the height of the printing bed is another factor to assess. The position in the z direction of the contouring requires a level of calibration for an optimal adhesion between each layer. According to the extruder's inner nozzle diameter, the printing height was adjusted accordingly. Such as a spiral vase, it was crucial to study the movement of the robotic arm when travelling from one curve to the following one to build a continuous trail to remove any

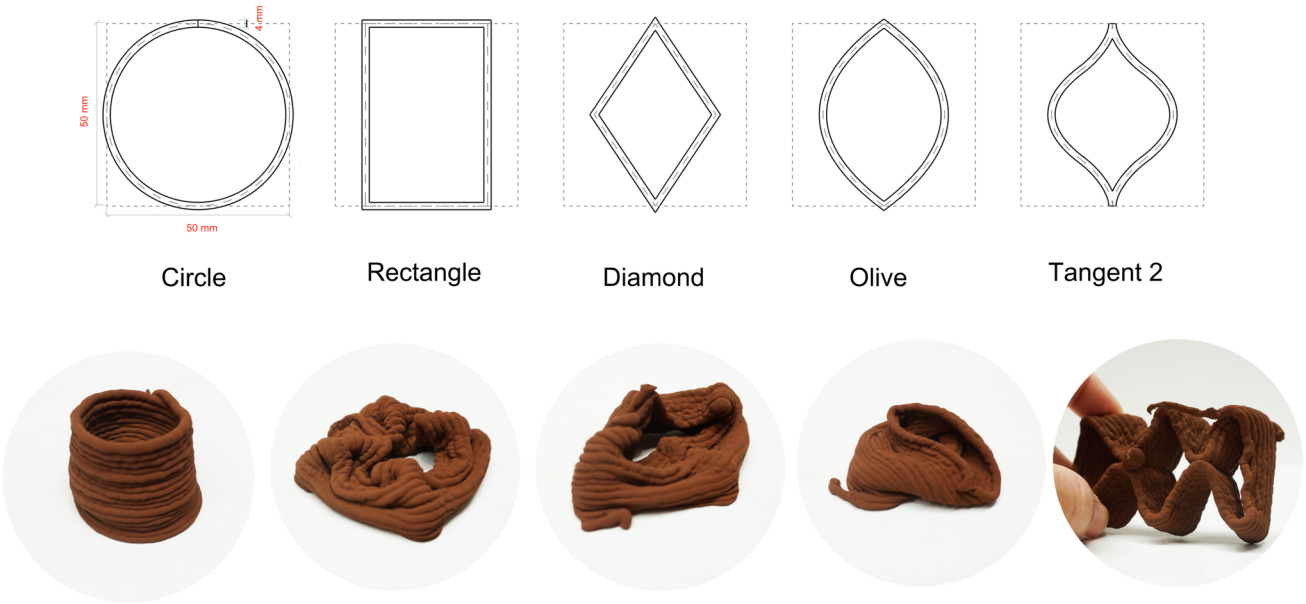

Fig. 10 Principal geometry testing (pictures & diagram by authors)

seams, resulting in printing at a faster rate and in achieving a smooth printed shape. Once the extruder was attached to the flange of the robotic arm and the stepping motor was rotating at a constant speed to extrude a homogenous paste onto an approachable printing bed, A series of tests regarding overhang, overlapping of layers, geometrical influences, infills were designed in Rhinoceros with the implementation of Grasshopper. This program was able to execute the desired printing shape and further evaluate the potentials and limitations of the process when printing with wood as an organic matter in a cold extrusion process.

The first challenge was to print a straight line at a certain height. Shown in Fig. 10, polygons like squares and diamonds tended to buckle above 30 mm of height and eventually collapse. Although material mass and the wall thickness demanded a balance to reach stability, circular geometries such as the circle in Fig. 10 have shown the highest stability. Therefore, another important factor to consider is a well-thought infill design to balance structural stability and density, and to be adjusted for each design. Wider layers and reduced layer heights offered more stability, although they increased the weight of the printed shape, reduced the resolution of the corners, and for straight walls they had the tendency to buckle and collapse. (Fig. 11). Also, the overlapping of a single toolpath was studied to connect all its edges for higher stability. In this study a nozzle diameter of 4 mm was used with 2 bar pressure and a print speed of 2000 mm/s. In grasshopper, the contouring of the geometry was established at a height of 3 mm with a 1 mm overlapping between its edges (Fig. 12). Lastly, to offer more

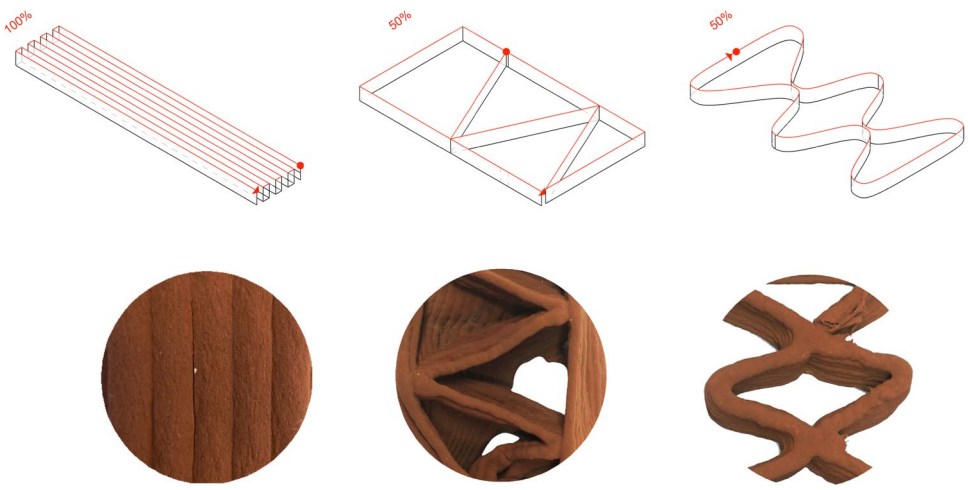

Fig. 11 Toolpath testing (pictures & diagrams by authors)

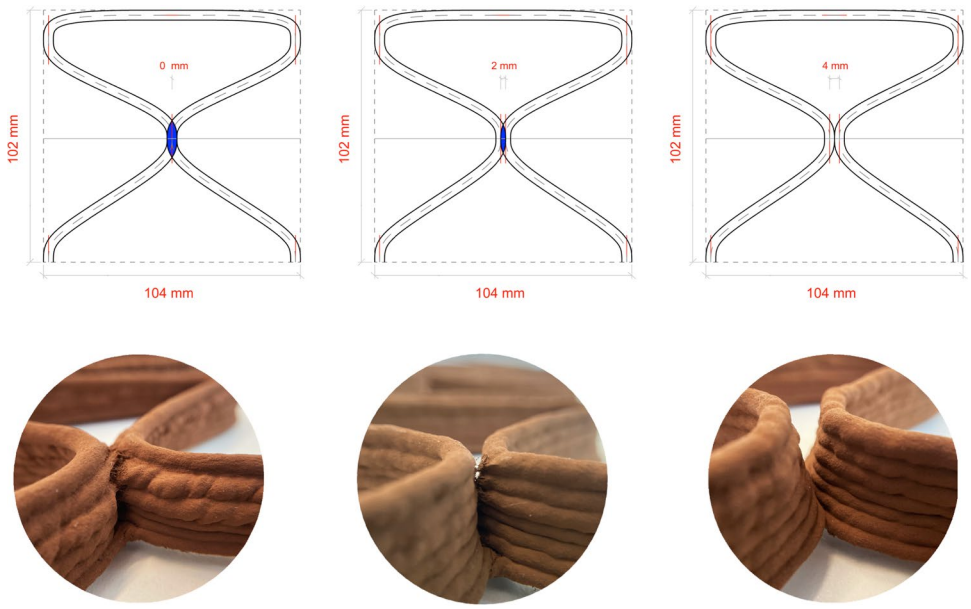

Fig. 12 Infill testing (pictures & diagrams by authors)

geometrical freedom to the desired printed shape overhangs were tested but failed at a 20-degree angle. Despite the failed tests, a customized infill design is necessary to provide adequate support without offering additional load on the inclined walls (Fig. 13). During these preliminary tests, it was observed that the overall drying time of the printed shape mainly depended on the temperature of the room and the amount of air that was in contact with its inner and outer cell. An unequal distribution of air influenced the shape of the final print. Also, after a curing time of 30 min, the material solidified enabling to print different components over themselves.

After studying the parameters to 3D print with methylcellulose in combination with lignin and cellulose, encountered challenges and potentials were the central drive to design and prototype a window frame and structural node as a proof of concept. During this phase, multiple aspects were identified when first printing the window frame (Fig. 14). From the previous tests, there was a clear restriction when printing under 10 cm. Another limitation regarding the maximum payload of the robotic arm which was 5 kg had to be considered. In consequence, with the custom-built holder weighting 4 kg, there was a small leverage of 800 g of material to be used in one single print. Consequently, the final design studied these limitations to understand the possibilities of printing a 180 × 180x100mm object. Thus, four different parts were printed and later combined. One printed section for one full cartridge would take 40 min to print 21 m of material. After a period of 7 days, the four separate elements were dry and shrunk by approximately 10 to 15 percent. The shrinkage of the material would cause printing issues for large objects as misalignments between the previous and ongoing printed layer would occur. Lastly, it was observed that the printed infill was too dense which

Fig. 13 Overhand testing (pictures & diagrams by authors)

Fig. 14 Window frame final prototype (picture by authors)

Fig. 15 Structural Node Final Prototype (picture by authors)

would have consequences on the drying time of the pieces as air does not have space to flow inside the cavities. Nevertheless, the curved infill system showcases the feasibility to perfectly connect and receive other components such as a gasket without additional sealants or adhesives.

An observed potential occurred as the material was drying at a fast rate, implying that objects could be printed monolithically, higher, faster, and stronger. Also, the slicing tool was studied to further print multiple parts over each other to potentially print a large object when emptying and refilling the cartridge. Also, an additive named bentonite as well as a reinforcement with long fibers named flax were studied and added to the next print to test the limits of the extruder when adding a drier, denser, and more viscous pastes. Based on the research of Chen et al. [63], in an additive manufacturing process, bentonite increased the material's viscosity and improves its extrudability. Flax, as a fiber reinforcement, was chosen due to its popular use in natural-fiber reinforced composites, availability, finest, and weight and yet reveals to have a high strength and stiffness [64]. In additive manufacturing, the potential of flax has been shown when blended with a thermoplastic matrix such as PLA to produce filaments [65]. Therefore, the structural node took another direction; an attempt for similar results were tested. The final extrusion with bentonite and flax showcased a strong and stiff material composition in comparison to the extrusions that were initially without them. The material was denser, heavier, and more resistant, allowing to successfully print a continuous and stable node of 100 mm heigh (Fig. 15).

Conclusion

The objective of this study was to develop a novel bio-based material from the building blocks of wood – cellulose, and lignin—potentially sourced from construction and agricultural residues and point toward an application using additive manufacturing. Raw materials and binding agents were explored, recipes defined, mechanical properties investigated, and printing parameters and limitations identified and documented.

A variety of 12 bio-based and synthetic binding agents were mixed with cellulose and lignin, analyzed, evaluated, and documented. Out of all alternatives, methylcellulose and wood glue demonstrated the highest potential, with the first one having the advantage of being natural and resulting in a bio-based formulation. The outcome was a homogeneous paste with moderate viscosity and adhesion, comparable to the clay commonly used in 3D printing, and capable of producing smooth and stable multi-layered extrusions with a syringe and a caulking gun. It did not present the same strength and stiffness as wood however, its mechanical properties placed it next to rigid polymer foams commonly used as insulation boards, performing better than other bio-based novel materials currently under development.

With the robotic arm and the extruder, the printability tests validated the potential of the methylcellulose-based mix as a feedstock for additive manufacturing and defined the limitations in terms of fabrication and material, conveyed through printing and design parameters. Extrusion speed, pressure, and nozzle diameter directly affected the smoothness and finishing of the printed parts. Curvilinear walls with a dense and winding infill and no overhangs proved stable, unlike linear structures built with single-line extrusions, which tended to buckle and collapse.

The results of the exploration steps guided the design and prototyping of two architectural elements – a fragment of a window frame and a structural node – to showcase potential applications of material and fabrication process in the built environment and challenge the limitations encountered. The window frame was fabricated in parts subsequently assembled. The node required further experimentation and improvements to the material to be extruded in one

continuous print. Flax fibers and bentonite were added to the paste to increase its stability and resulted in a successful monolithic print of 100 mm in height. The mechanical properties of the enhanced mix were not assessed, although they were seemingly improved, indicating a direction for further research.

Overall the lack of mechanical properties comparable to wood, carbon fiber reinforced polymers, or steel hinder its use in stiffness and strength-driven applications. Nonetheless, this study succeeded at developing a bio-based and wood-based feedstock and applying it in a liquid deposition additive manufacturing process. Further research is necessary to enhance the mechanical properties and investigate its potential uses in the construction industry.

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Data Availability This study succeeded at developing preliminary datasets (https://repository.tudelft.nl/islandora/object/uuid:9e0e0d92-e1b3-42dd-b2ca-1a105ec571de), producing a biobased and wood-based feedstock.

Declarations

Conflict of interest Conflict of interest on behalf of all authors, the corresponding author states that there is no conflict of interest.

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