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Effect of wood content in FDM filament on properties of 3D printed parts

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ABSTRACT

The effect of wood content in 3D printing materials on the properties of 3D printed parts was investigated. Six filaments using polylactic acid (PLA) with varying loading levels of wood particles from 0% to 50% by weight were produced and used for 3D printing. The density of the filaments and 3D printed parts used in this study slightly decreased with increasing wood content. The tensile strength of the filaments increased from 55 MPa to 57 MPa with an addition of 10% wood, but decreased with higher levels of wood content to 30 MPa for filaments with 50% wood content. The surface of the parts printed from the filament without the addition of wood was smoother and the printed part had no voids within the structure. With increasing wood content the surface becomes rougher, more voids were present, and had visible clusters of wood particles (due to wood particle clustering and clogging in the printer nozzle). Higher wood content in 3D printed parts decreased the storage modulus. measured with torsional loading on a rheometer, but did not change the glass transition temperature.

1. Introduction

Additive manufacturing, commonly known as 3D printing, has greatly developed in the last decade. It has several advantages compared to subtractive manufacturing, especially in its ability to produce complex shapes and products without special tools or moulds, making it ideal for developing prototypes or customized products [1]. The efficient use of raw materials in 3D printing is leading to low or zero waste manufacturing [2], which is becoming increasingly important.

There has been major interest in biodegradable 3D printing which includes the development and testing of eco-friendly and recycled materials including wood, cellulose, sugars, and lignin [1,3]. There has also been interest in fibre, particle, and nanocomposite reinforced materials [4], and in the last few years several materials appeared on the market containing wood particles. Several techniques for 3D printing with wood have been used including fused deposition modelling (FDM), injection powder printing [5], and liquid/paste deposition modelling [6].

The wood-plastic composite (WPC) industry has also utilised natural materials in their products. WPCs are composite materials made of recycled and/or virgin plastic and wood particles. They are used for outdoor decking, railings, fences, landscaping timbers, cladding and siding, park benches, moulding and trim, window and door frames, and indoor furniture [7]. WPC and 3D printing both extrude molten thermoplastic polymers compounded with wood particles. WPC processing technology extrudes the mixture through a die and into a certain profile

with a fixed and relatively simple shape.FDM extrudes the mixture through a round nozzle and forms shapes with controlled deposition of materials in layers. There are also differences in processing parameters concerning the equipment, pressures during extrusion, and cooling/hardening conditions after extrusion. The low price of residual wood particles is one reason they are used in WPCs, which also limits the amount of more costly thermoplastic polymers. The use of natural materials like wood helps to lower the usage of petroleum based plastics and reduce environmental impacts [7]. These benefits also extend to 3D printed materials.

With the right combination of polymer, filler, and additives a wide variety of performance levels in WPC and 3D printed materials can be achieved. For example, researchers indicate that beech sawdust can act as a reinforcement in the case of flexural stress and tensile strength. The addition of sawdust reduces the WPC density and despite the increased brittleness creates a convenient and low-cost alternative to composites reinforced with synthetic fillers like glass fibres [8]. The effect of the wood on WPC properties depends on several factors from wood particle properties, size distribution, and compatibility with the thermoplastic [7]. The use of wood fillers in thermoplastics is likely to increase with development of improved compounding techniques and new coupling agents that permit usage of high filler/reinforcement content [7].

WPCs can be used as 3D printing material on a modified 3D printer. The key is to grind wood into particles of the appropriate size fineness and texture allowing it to be mixed into a polymer [1]. In 3D printing the size of the wood particles is also limited by the printing nozzle

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diameter. The most commonly used polymers for 3D printing are: acrylnitrile butadiene styrene (ABS), polylactic acid (PLA), and different blends such as PLA with polyhydroxyalkanoates (PHA).

Filaments from PLA biocomposites can be utilized in 3D printing due to good interfacial bonding between the biomaterial and the PLA matrix. Good interfacial bonding has been found with various natural materials like dried distillers grain with solubles, Paulownia wood, and Osage orange wood [9]. The research showed that tensile strength values of the biocomposites were considerably lower than neat PLA [9], but the Young's modulus (MOE), and elongation at break (ELO) were comparable to neat PLA in most cases. The reduction of tensile strength values in biocomposites is typical due to poor interfacial bonding between the hydrophobic PLA and hydrophilic natural materials. In these cases, additives and processing techniques should be used to improve material properties [9].

The compatibility between the polymer and wood components of the composite can be improved using either physical or chemical modification of the polymer or wood particles or by using coupling agents. The final properties of such composites largely depend on the compounding process and processing conditions [10].

The results of previous studies show that mechanical properties of 3D printed parts depend upon the printing pattern and build style. The samples printed with a solid build style exhibit higher storage modulus than samples with mesh structures [11]. High ambient temperatures can also decrease the mechanical properties of the printed parts due to of the polymer softening. In samples printed with mesh structures, empty spaces can be present with more or less sharp crack tip where high stress concentrations can be present, leading to fracture [12] or can lower the dynamic bending strength [13].

Natural polymers derived from different plants can also be used in 3D printing [14]. Printed materials using these polymers showed lower mechanical properties compared to conventional commercial plastics, but are often adequate for the desired application and can be increased with the addition of additives, plasticizers, or with modification of manufacturing parameters.

When using fibres in composites, their orientation is important for the final product properties. In FDM 3D printing, wood-fibre orientation follows the printing orientation of the filament, while inter filament and interlayer interactions could be considered as the weakest link of the printed material. Some specific properties of wood also affect wood plastic composites like a high swelling ratio due to their anisotropic hygroscopic properties [15]. The hygroscopic properties could be turned into an advantage in the design of new types of passive hygromorph products, where we could generate a programmable moisture-actuated functionality for biocomposites, by following the 4D printing concept [15].

Over the past two decades, the thermoplastic industry has been attempting to decrease the dependence on petroleum-based fuels and products due to environmental awareness and international demand for green technology. The use of natural fibers instead of traditional reinforcement materials such as glass fibers, carbon, and talc provides several advantages including their low density, good thermal insulation and mechanical properties, reduced tool wear, unlimited availability, low price, and biodegradability [16]. Natural fibers also offer economical and environmental advantages compared with traditional inorganic reinforcements and fillers.

In this study, the different amounts of the wood flour as reinforcing filler was incorporated into thermoplastic filament. Some applications of 3D filaments needs specific mechanical and thermal properties as compared to the traditional petrolum filaments such as polypropylene, polyethlyne, poly(lactic) acid. In addition, environmental consciousness and increasing filament prices force the filament producers and consumers to environmentally friendly materials. Based on the extensive literature search, there is limited information on the use of wood in the 3D filaments. In this study, the optimum wood flour content was determined based on the physical, mechanical, and rheological

properties of the 3D printed composites.

2. Materials

Five 3D printing filaments with different wood content and two commonly used commercial filaments (without wood particles) were used. Wood particles used in the filaments were prepared by milling beech wood (*Fagus sylvatica* L.) in a laboratory mill (Retsch ZM 200). The wood particles were then sieved through different meshes and only particles that went through the mesh with an opening of 0.237 mm were used to produce filaments. The particle size distribution and aspect ratio of particles were not measured and will be part of further research since they can significantly influence the properties of composite.

The thermoplastic used was PLA Ingeo™ 2003D (NatureWorks, Blair, NE) in granulated form. Granules and wood particles were dried in a laboratory dryer before preparing the mixtures. Five different filaments were prepared with different wood content and one control filament: 0%, 10%, 20%, 30%, 40% and 50% wood content by weight.

The wood particles and PLA granules were first compounded and pelletized for subsequent processing. The wood-PLA pellets were then extruded using a single-screw filament Noztek-pro extruder (Noztek, *Shoreham, UK*) to produce the filament. The filament thickness was set to 1.75 mm, but the final thickness varied between 1.45 mm and 1.75 mm.

The two commercially available filaments without wood particles were Z-ABS filament (Zortrax S.A., Poland) and PLA PrimaValue filament (3D Prima, Sweden). Physical, mechanical, and rheologic properties of these two filaments were tested with preliminary research using the same methods as other filaments and served as a comparison to our filaments (Table 1).

A Zortrax M200 3D printer (Zortrax, Poland) was used to print our test samples. The printing temperature for ABS was 275 $^{\circ}$ C and for PLA filaments, 230 $^{\circ}$ C. The nozzle had a 0.4 mm opening and the printing layer thickness was set to 0.19 mm. The same 3D model with dimensions of 40 mm x 12 mm x 4 mm was printed with all filaments. The model was made from solid layers on the bottom, top, and sides, but the inside was a mesh structure (solid infill setting: square size 1.25 mm and line thickness is 0.4 mm).

3. Methods

The density of the filament and 3D printed parts was determined by measuring dimensions and mass. Five samples were measured for each material and average values were determined.

The tensile strength of the filaments was tested on a Zwick-Roel Z005 universal testing machine. Five samples were measured for each material and average values were determined. A test speed of $10\,\mathrm{mm/min}$ was used.

A digital camera and microscope were used to capture images of the surface and edges of 3D printed parts and filaments at different magnifications.

A TA Instruments Ares-G2 rheometer was used for determination of rheological properties of 3D printed parts. A torsion fixture was used to

 Table 1

 Properties of commercial filaments used in this research.

	PLA	ABS
Filament density (g/cm³)	1.25	1.06
3D printed parts density (g/cm ³)	0.70	0.67
E modulus tensile (GPa)	3.72	2.48
Tensile strength (MPa)	55.3	42.7
Linear viscoelastic region limit strain (%)	0.63	0.79
Max storage modulus (GPa)	0.67	0.45
Glass transition temperature (°C)	66	119

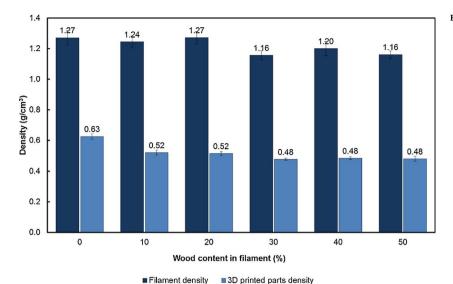


Fig. 1. Filament and 3D printed parts density.

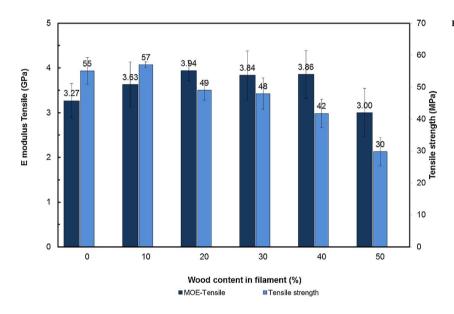


Fig. 2. Filament tensile strength and MOE.

clamp the samples into the rheometer and oscillation torsion loading was used. Two tests were done on each sample: amplitude sweep and a temperature ramp test. Two samples were tested for each material.

Amplitude sweep was used to measure the reaction of the material to loading with different amplitudes. The strain increased in this test from $1\times 10^{-3}\%$ to the end of the linear viscoelastic region (LVR) of the material. The end of LVR was determined as a 5% drop of initial storage modulus.

LVR is defined as a region corresponding to the strain varying linearly with stress. The response (strain) is directly proportional to the mechanical input (stress), the polymer is not altered, and the response reflects the polymer structure and organisation [17]. The proportional limit is the point at which the relationship between stress and strain is no longer linear. Points beyond the proportional limit can cause irreversible physical changes in the material [18]. If the amplitude is properly selected within the LVR of the material being used, the material structure remains intact and is not permanently deformed [19].

In addition to knowing viscoelastic properties, mechanical analysis can provide information about material compatibility with other materials. For example, the compatibility between a wood surface and adhesives or paints [17].

The strain level used in further research should be in the LVR of the material and also as large as possible to achieve the highest response of

material to loading. Based on the results from the amplitude sweeps we selected a strain rate of 0.4% for subsequent temperature ramp tests. This strain rate was selected for all materials near the end of the linear region and gave good results in further tests.

The temperature ramp test was conducted on the same samples with a temperature range of 30 $^{\circ}\text{C}{-}110\,^{\circ}\text{C}$ and with a heating rate of 10 $^{\circ}\text{C}/$ min. The oscillation loading strain was 0.4% and loading frequency was 1 Hz. During temperature ramp test we measure response of the material to loading at increasing ambient temperatures.

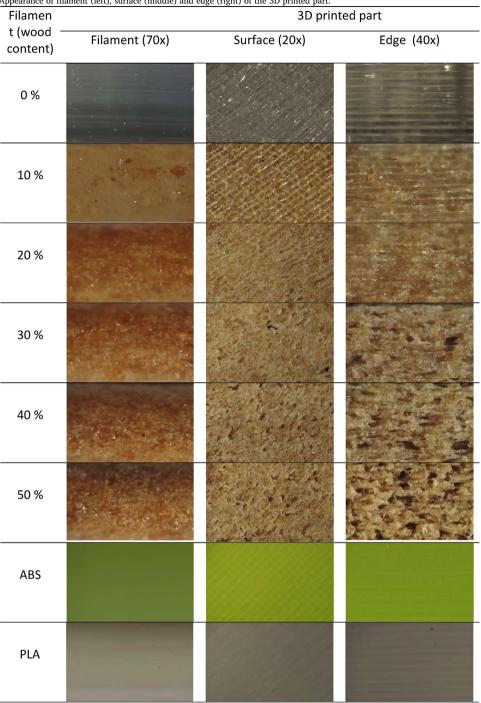
The glass transition temperature can be determined from the tan δ curve. Tan δ is the ratio of the loss modulus to the storage modulus and is a measure of the energy dissipation of a material. The peak of the tan δ curve identifies the temperature of glass transition temperature.

4. Results and discussion

4.1. Properties of filaments and 3D printed parts

The density of the filaments decreased with increasing wood content (Fig. 1). This was expected since the density of PLA was 1.24 g/cm³ (manufacturer data) and the density of beech wood is approximately 0.65 g/cm³. So the addition of wood should decrease the density of filaments. The values are comparable to commercial filaments, which

Table 2
Appearance of filament (left), surface (middle) and edge (right) of the 3D printed part.



have $1.25\,\mathrm{g/cm^3}$ (PLA filament) and $1.06\,\mathrm{g/cm^3}$ (ABS filament) (Table 1).

The structure of the 3D printed parts was the same with three full layers on the top/bottom and sides, and a mesh structure in the middle. However, the mass and density of the 3D printed parts varied. The highest density printed parts used the filament without an addition of wood $(0.63~{\rm g/cm^3})$. The density decreased in the samples with 10% and 20% wood content, to $0.52~{\rm g/cm^3}$. The lowest density samples were those with 30% to 50% wood with a density of $0.48~{\rm g/cm^3}$. Samples with a wood content of 10% and 20% decreased the density by 17% and samples with 30% to 50% wood had a 24% lower density.

The reason for these differences is the lower density of wood, but also in the total quantity of filament material (wood and polymer) present in the 3D printed parts. Filaments with higher content of wood did not flow evenly through the nozzle and often clogged it resulting in poorly printed parts that were not evenly filled with material. There were also slight differences in the filament diameter, which changed the amount of material extruded to printed parts. The amount of the extruded material at selected printing speed influences the imprint into previously deposited material and thus influences the connection (contact area) between printed lines and layers and consequently the strength of printed part. The interface between wood particles and PLA matrix was not investigated in this research, but different additives can be used to improve bonding interface and processing.

The tensile strength of the filaments increased from 55 MPa to 57 MPa with 10% wood addition (Fig. 2), but decreased with higher

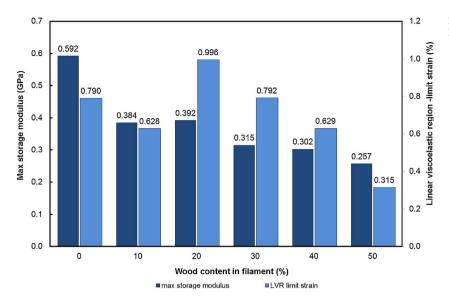


Fig. 3. Max storage modulus and strain at linear viscoelastic region limit

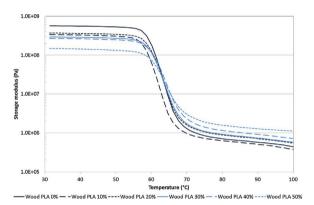


Fig. 4. Effect of temperature on storage modulus of different 3D printing materials.

wood content to 30 MPa for the filament with 50% wood content. The elastic modulus of the filaments have a slightly different trend. The control filament without wood had a modulus of elasticity (MOE) value of 3.27 GPa. This value increased with the addition of 20% wood content to 3.94 GPa. Further addition of wood resulted in lower MOE values and dropped to 3.00 GPa for samples with 50% wood content.

Small increases in the MOE and tensile strength values were measured with lower additions of wood, but these values decreased with higher loading levels. With low levels of wood content, the wood particles can act as reinforcement, but at higher loading levels, the polymer cannot fully encapsulate the particles, leading to poor bonding and limited load transfer.

The wood particles used in this study had a relatively small aspect ratio and could not contribute to the mechanical properties in such a way as longer fibres/particles would. The nozzle diameter restricts the size of wood particles being used and if larger particles were used, more clogging would occur.

4.2. Visual differences of surface of 3D printed parts

The unfilled PLA granules had no added colour and the 3D printed parts from this filament were almost transparent (Table 2). With the addition of wood particles, the filament and 3D printed parts become less transparent and the surface became similar to the surface of medium density fibreboards (MDF). The surface created from PLA without the addition of wood was smooth and had no voids, but with increasing wood content it became rougher, more porous, and had visible clusters of wood particles. Dark spots present on the edges of

printed parts with 30% to 50% wood particles (Table 2) are voids where material was not deposited due to clogging and irregular flow through the nozzle. Additives serving as lubricants, more precise temperature regulation, and extrusion speed control would be needed to improve the surface quality. This irregular surface also contributed to lower mechanical properties due to poorly bonded wood particles which ineffectively transfer load.

4.3. Rheological properties

The samples with 20% wood content showed the highest LVR limit strain (0.996%) (Fig. 3) and the samples with 50% wood content had the lowest LVR limit strain

(0.315%). The remaining LVR limit strain values are comparable to printed parts from commercial PLA (0.63%) and ABS (0.79%) (Table 1).

The storage modulus (G') is the measure of the elastic behaviour of the sample. G'of printed parts from commercially available PLA filament was the highest at 0.67 GPa. Parts printed from PLA filaments produced in this study with no wood showed a smaller storage modulus (0.59 GPa). The addition of wood particles decreased G'. The addition of 10% to 20% of wood decreased the G' by 33%. Increasing the wood content decreased G' at a slower rate reaching a 56% drop in G' for samples made with 50% wood content (0.26 GPa). The reason for the lower storage modulus is in a smaller diameter filament resulting in less material in the 3D printed parts, and non-fused material due to clogging and clustering of wood particles. The printed ABS parts showed a lower modulus (0.45 GPa) (Table 1) compared to PLA, which was expected [20], but still higher than the parts printed with filaments filled with wood.

The temperature sweep test (Fig. 4) shows the behaviour of the 3D printed parts due to heating from 30 $^{\circ}\text{C}$ to $110\,^{\circ}\text{C}$. The material keeps its strength until it reaches the glass transition temperature, where the storage modulus rapidly decreases for several decades. The 3D printed parts then lose their strength and become soft, deforming with increasing temperature.

The printed ABS parts soften at the highest temperatures, above $100\,^{\circ}\text{C}$ (Table 1). Parts printed from ABS could be used at temperatures up to $90\,^{\circ}\text{C}$ due to them holding their strength at elevated temperatures. Parts printed from PLA started to soften at $60\,^{\circ}\text{C}$ (Fig. 4). Parts with wood added to them also softened near $60\,^{\circ}\text{C}$, but have a lower storage modulus before their glass transition temperature. The parts with higher content of wood showed higher storage modulus after the glass transition temperature than pure PLA. The wood particles probably maintained a part of this storage modulus.

The measurements showed that the glass transition temperature did not change with addition of wood and was between 65 °C and 66 °C. The primary phase of the structure is the PLA polymer matrix which softened at 66 °C and the smaller wood phase has little effect on the material properties during heating.

5. Conclusions

The results showed that wood particles can be used as a component in materials for FDM 3D printing. The wood particles added to filaments decreased the density of the filaments and 3D printed parts due to the lower density of wood compared to PLA.

The tensile strength of the filaments slightly increased from $55\,\mathrm{MPa}$ to $57\,\mathrm{MPa}$ with a 10% addition of wood, but decreased with higher wood content to $30\,\mathrm{MPa}$ for filaments with 50% wood content. Lower levels of added wood particles slightly reinforced the filament until 10%, but with higher levels of wood added, the tensile strength decreased. The results are not statistically significant due to small samples size and large standard deviation.

The surfaces of printed parts from PLA with no added wood were smooth and had no voids, but with increasing wood content it became rougher, more porous, and had visible clusters of wood particles. These non-solid/fused structures contributed to lower mechanical properties of 3D printed parts. The addition of wood particles decreased the torsion storage modulus of 3D printed parts.

The measurements showed that the glass transition temperature did not change significantly with the addition of wood and was between 65 °C and 66 °C for filaments produced in this study, since the main matrix (PLA) material was the same for all filaments.

Further research with different wood particles or fibres (wood species, size of particles, length to diameter ratio, particle surface treatment), and printing parameters (temperature, nozzle diameter, precise feeding speed regulation) needs to be done to optimize the printing of 3D parts that would be comparable in performance and properties to those using commercial filaments without wood.

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