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Investigation into the material properties of wooden composite structures with in-situ fibre reinforcement using additive manufacturing

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ABSTRACT

In contrast to subtractive manufacturing techniques, additive manufacturing processes are known for their high efficiency in regards to utilisation of feedstock. However the various polymer, metallic and composite feedstocks used within additive manufacturing are mainly derived from energy consuming, inefficient methods, often originating from non-sustainable sources. This work explores the mechanical properties of additively manufactured composite structures fabricated from recycled sustainable wood waste with the aim of enhancing mechanical properties through glass fibre reinforcement.

In the first instance, samples were formed by pouring formulation of wood waste (wood flour) and thermosetting binder (urea formaldehyde), with and without glass fibres, into a mould. The same formulations were used to additively manufacture samples via a layered deposition technique. Samples manufactured using each technique were cured and subsequently tested for their mechanical properties. Additively manufactured samples had superior mechanical properties, with up to 73% increase in tensile strength compared to moulded composites due to a densification of feedstock/paste and fibre in-situ directional alignment.

1. Introduction

Additive Manufacturing (AM), also known as 3D printing, is a layer by layer fabrication of 3D objects from a digital model. During the process, material is laid down in individual layers; each layer is bonded/fused together through various techniques (sintering, melting, curing, chemical reactions etc.) [1]. This differs from subtractive manufacturing techniques such as machining that remove material which is considered to be an inefficient process in regards to material utilisation. AM technologies make it possible to build a large range of functional components with complex geometries which may be difficult, or even impossible, to achieve using conventional methods. Furthermore, manufacturing development cycles can be shortened when using AM, thereby reducing productions costs [2]. AM technologies have been involved in various applications in areas such as aerospace [3], automotive [4], artistic design [5] and biomedicine [6]. Materials commonly used in AM are often plastic [7] or metals based [8]. Although these additive processes are known for their low material waste during part manufacture, the creation of this feedstock is often through inefficient means. In contrast, there are currently only a few commercially available materials that are created from natural feedstock [9].

The AM of wood waste presents an opportunity to create 3D components from a cheap and sustainable source with limited material losses during processing. Wood flour is a typical wood waste and is processed commercially from post-industrial processes such as chips, sawdust and planar shavings. Unlike the larger sized chips, fibres and flakes, which are typically used in combination with thermosetting adhesive resins to produce wood panel products, wood flour is commonly used as a reinforcing filler in thermoplastic composite materials [10–12]. Wood-thermoplastic composites have become a widely recognised commercial product in construction, furniture and other consumer applications [13]. Wood-plastic composite components may be produced from a variety of different techniques such as injection-moulding [14], compression moulding [15], and extrusion [16]. The extrusion technique allows for net-shape components to be manufactured

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additively. There are several patents claiming to manufacture wood products via AM. Here, combinations of wood flour/fibres and variety of plastics such as acrylonitrile butadiene styrene [9,17], polylactic acid [9,17], and polypropylene [9,18] are used as the feedstock materials, together with a range of additives such as coupling agents, plastisizers, dispersants and lubricants. A further patent describes a method to produce wood products by which a binding agent is deposited via an ink jet printer head onto layers of wood powder [19]. However, the properties of the printed product are not reported.

In addition to thermoplastics, there are many other wood binders such as thermosetting resins and inorganic cements which are used in the production of wood composite materials [20,21]. Thermosetting adhesives have advantages over thermoplastic adhesives as they offer enhanced product qualities such as improved temperature resistance, resistance to deformation and superior mechanical properties [22–24]. Consequently, they are the preferred adhesives in the production of materials for structural applications such as wood panel products (e.g plywood, strandboard and fibreboard). AM of wood using adhesives other than thermoplastics has been reported [25]. However, there are limited studies which detail the methods used and which examine the mechanical properties of the printed product [26]. One such study describes how thin layers of wood chips/inorganic binder are bonded together to produce a solid of desired shape [21]. Bonding and hardening is brought about by the use of aerolised water as an activator on each layer. Binders investigated included gypsum, sodium silicate and a variety of cements. Investigators [27] have combined beech wood pulp with a starch binder (hyroxypropyl) to form a paste that was extruded/deposited with a syringe to form 3D wooden structures. However, due to the binder used, samples were very weak (maximum tensile stress of 3.5 MPa) and held similar properties to that of wood cements. Another work extruding wood pulp [28], used varying proportions of beech wood mixed with polyvinyl acetate and urea formaldehyde. It was found that the bending strength and stiffness of formed 3D parts improved when the urea formaldehyde binder was used, however properties were not affected by the amount of wood pulp used within the composition. This and other studies [29] conclude that material properties need to be improved in order for additively manufactured wooden structures to be suited for large structural applications.

Although the products are not, as yet, of sufficient mechanical strength suitable for many engineering applications, research has demonstrated that wood/adhesive composites can be produced by additive manufacturing. Furthermore, additive manufacturing technologies offer the possibility of inducing the alignment of fibrous materials within a product to enhance mechanical strength. Indeed, Compton and Lewis [30] reported the alignment of high aspect ratio fibres in epoxy resin using a 3D printing method, yielding improved mechanical properties along the printing direction. In this paper, we explore the applicability of an additive manufacturing technique to exploit potential fibre alignment within wood-thermosetting binder composites in order to improve mechanical properties.

2. Methodology

The creation of 3D printed components from wood flour and thermosetting binders was achieved through extruding these formulations, in a form of a paste, through a fine nozzle. Wood flour was selected as the wood component of the composites as it is considered a cheap and sustainable source of wood waste. Furthermore, the relatively small size of wood flour particles, compared to other wood wastes, make it suitable for 3D printing via extrusion through a nozzle. Urea formaldehyde was selected as the binder material. It is a traditional wood glue, commonly used as a thermosetting resin in the manufacture of panel products [20]. It was anticipated that the addition of strength enhancing fibres to the formulations would improve the mechanical properties of the composites [31] and, therefore, the effect of incorporating glass fibres into the composites was investigated. The mechanical properties of these 3D printed composites were subsequently examined and compared to non-printed, moulded composites prepared using the same formulations.

2.1. Materials

Wood flour waste feedstock was purchased from Eden Products Ltd, Middlewich, Cheshire, UK. This product (EPWF 110) is a waste wood from European white softwood (South Germany). The particle size distribution of the wood flour fibres was obtained using a sieving technique (Retsch AS200 sieve shaker), yielding a d_{50} (median) size value of 75 μm (it should be noted that this value obtained via sieving refers to the shortest physical dimension of the wood flour fibres). The wood flour samples were stored at a constant temperature of 21 °C and the moisture content was determined to be 14%. This value is within the accepted level for use with wood adhesives (<15%) and, therefore, adhesive performance would not be compromised. Urea formaldehyde (CASCORIT 1205) and hardener 2545 were both purchased form Glues Direct, UK. The pot life of the urea formaldehyde/hardener system is 8 h at room temperature and pressure, allowing sufficient time for formulation preparation and 3D printing of the pastes. Glass fibres (Vitrostrand), of approximately 100 μm in length, were purchased from East Coast Fibreglass, UK.

2.2. Composite manufacture

2.2.1. Additively manufactured composites

2.2.1.1. Preparation of formulations. A formulation of 13%wt/wt wood flour in urea formaldehyde was used. This had a viscosity that was low enough to be extruded/deposited through the nozzle easily, but also held a suitably high level of viscosity, enabling the dispensed product to maintain its shape prior to curing. The urea formaldehyde and hardener were mixed thoroughly at room temperature at a mixing ratio of 100 pbw urea formaldehyde: 20 pbw hardener. After degassing, the wood flour was added to the adhesive at room temperature and mixed thoroughly by hand using a wooden spatula.

Secondly, a formulation consisting of 8.8%wt/wt wood flour and 10%wt/wt glass fibres in urea formaldehyde was prepared. As with the non-glass fibre paste, this paste could be extruded/deposited easily through the nozzle whilst maintaining its shape before curing. Table 1 details the composite formulations in terms of weight fraction, volume fraction and density.

2.2.1.2. Additive manufacturing via extrusion/deposition. Printing via extrusion was carried out at room temperature (21 °C) using a Fisnar robot 7400; a 3-axis robot with a working area of 400 × 400 × 100 mm. A schematic of the process is shown in Fig. 1. The operating parameters (printing co-ordinates, printing line speed of 15 mm/s) were controlled using Smart Robot Edit software (Fisnar) running on a PC. The formulations were prepared and placed in a syringe barrel. The barrel was then attached to a pneumatic general purpose dispenser (Fisnar JB113N), connected to a compressed air supply (20psi). The dispenser was interfaced with the robot and controlled the dispensing pressure. The syringe barrel containing the paste was then placed in the barrel holder on the robot and a 1.6 mm diameter nozzle attached to the bottom of the barrel. Rectangular samples (80 mm × 10 mm x 2 mm) were

Table 1
Composite formulations (UF = urea formaldehyde) and theoretical density assuming no voids.

Composite	Composite components	Component density (g/cm^3)	Weight fraction	Volume fraction	Theoretical composite density (g/cm^3)
Wood flour/UF only	wood	1.55	0.13	0.11	1.32
	UF	1.30	0.87	0.89	
Wood flour/UF with glass fibres	wood	1.55	0.09	0.08	1.38
	glass	2.54	0.10	0.05	
	UF	1.30	0.81	0.87	

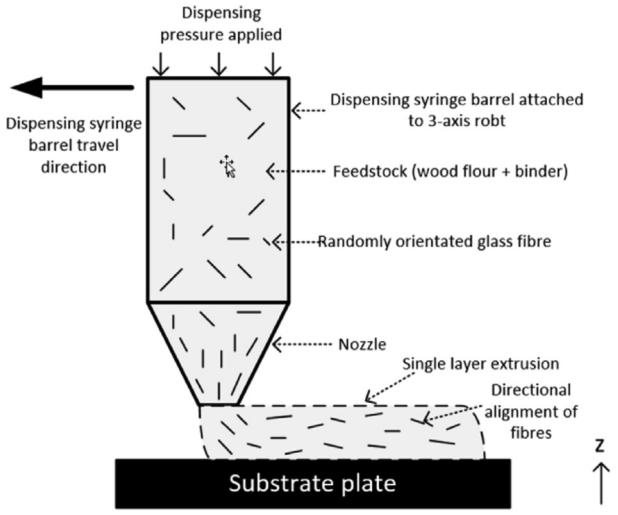

Fig. 1. Schematic of the extrusion/deposition based additive manufacturing technique and hypothesis of glass fibre alignment.

printed for flexural three point bend tests and 'dog bone' samples (30 mm × 10 mm x 2 mm gauge section) were printed for tensile strength testing. Both these samples consisted of two layers. As can be seen in Fig. 1, due to the high aspect ratio of fibres it is hypothesised that any fibres present within the feedstock mixture will undergo directional alignment as the material is extruded/deposited through a nozzle with a progressive reduction in diameter. Alignment of particles with high aspect ratios using printing techniques has previously been reported to give materials enhanced mechanical properties along the printing direction [30,32]. Subsequent SEM analysis will confirm position and any alignment of fibres within the printed structure.

2.2.1.3. Product curing. After printing, the products were cured in an oven using the following cure cycle (with a 0.1°C/min ramp between each point in the cycle): 50 °C for 1hr, 60 °C for 1hr, 70 °C for 1hr, 80 °C for 1 h, 90 °C for 1 h, 100 °C for 1hr and 110 °C for 2hr.

2.2.2. Preparation of non-printed composites

Additionally, non-printed, moulded composites of the aforementioned formulations were also prepared for comparison of mechanical properties with printed composites. Here, the pastes were simply placed into rectangular-shaped moulds (80 mm × 10 mm x 2 mm) for flexural testing and dog-bone shaped moulds (30 mm × 10 mm x 2 mm gauge section) for tensile testing and cured in an oven using the same cure cycle as that for the printed composites.

2.3. Composite analysis

2.3.1. Mechanical property testing

The cured composites were subsequently tested for their mechanical properties. Flexural and tensile data were directly tested and calculated independent from each other according to ISO standards. Three point bend testing was used to obtain values of the flexural modulus and flexural strength. Tests were carried out using a TA500 Texture Analyser with a 500N load cell, and five tests were performed for each sample type. Flexural testing was carried out with reference to: Plastics – Determination of flexural properties (ISO 178:2010) [33]. Test speed was carried out at 1 mm/min in accordance with this standard.

Tensile strength testing was also carried out using the same TA500 Texture Analyser. The ultimate tensile strength is the maximum stress sustained by the sample during the test before failure. Five tests were performed for each sample type. Tensile testing was carried out with reference to Plastics – Determination of tensile properties (ISO 527–2:2012). Part 2: Test conditions for moulding and extrusion plastics (specimens of type 1BA) [34]. This standard was chosen as the most appropriate to allow comparison of the printed vs moulded samples.

The density of each of the composites was calculated by simply measuring the volume and mass of the rectangular samples prior to flexural testing.

2.3.2. Composite structure

Image analysis of cross-sectioned samples was undertaken using a FEI Sirion Inspect F50 Scanning Electron Microscope (SEM). Samples were cut, grounded, polished and dried according to standard procedures. The wood samples were mounted onto specimen stubs using conductive adhesive, and coated with gold prior to analysis.

3. Results and discussion

The mechanical properties of both printed and non-printed wood composites are presented in Table 2, Fig. 2 and Fig. 3. From the gradients of the flexural stress-strain curves, it was evident that the printed samples demonstrated a higher stiffness than that of the non-printed samples and had a higher flexural strength. The mechanical properties of the composites are also compared to literature values of wood and wood panel products (Table 3) [35]. The flexural modulus of printed composites had increased to a value greater than that of hardboard and approaching that of plywood. The printed samples possess a considerably higher tensile strength compared to non-printed samples. The tensile strength of the printed samples was now much greater than that of particleboard with the values approaching that of hardboard (see Table 3).

These superior mechanical properties are due to a densification of the paste as it is extruded/deposited through the nozzle. Indeed, density measurements (Table 2) show that the cured printed samples have a higher density compared to cured non-printed samples. Extrusion removes microscopic air bubbles in the feedstock during the process as it passes through the extruding nozzle under pressure, thus making the product denser and improving both flexural and tensile strength. Furthermore, the voids content, calculated from experimental density and the theoretical density (Table 1) is significantly less for the printed composites (7.3%)

Table 2
Mechanical properties of printed composites compared to non-printed composites.

Composite		Density (g/cm^3)	Voids content (%)	Flexural modulus/GPa	Flexural strength/MPa	Tensile strength/MPa
Wood flour only	Non-printed	1.16 ± 0.01	12.3 ± 0.88	5.4 ± 0.4	44 ± 3	9.7 ± 1.0
	Printed	1.22 ± 0.02	7.30 ± 1.21	6.8 ± 0.6	57 ± 5	17 ± 1.0
Wood flour and glass fibres	Non-printed	1.21 ± 0.01	12.3 ± 1.05	5.9 ± 0.7	38 ± 5	16 ± 1.0
	Printed	1.31 ± 0.01	4.90 ± 0.46	8.7 ± 0.2	58 ± 2	22 ± 2.0

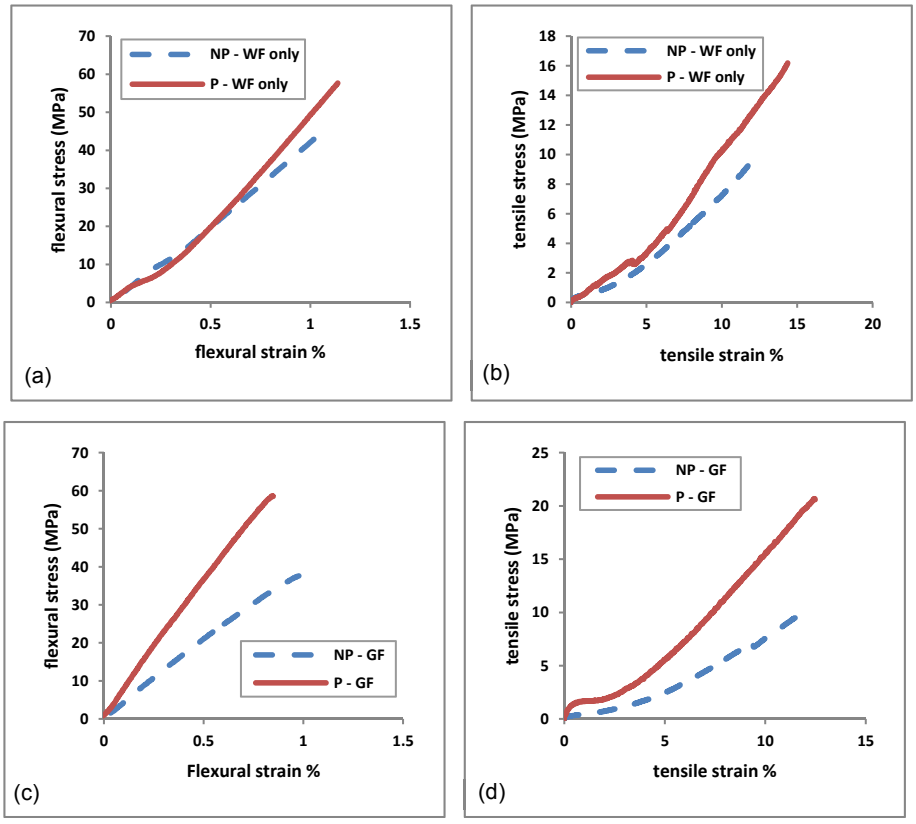

Fig. 2. Comparison of mechanical properties of printed and non-printed products: (a) flexural and (b) tensile stress strain curves for urea formaldehyde-wood flour only samples; (c) flexural and (d) tensile stress-strain curves for urea formaldehyde - wood flour - glass fibre samples. P = printed, NP = non-printed, WF = wood flour, GF = with glass fibres present.

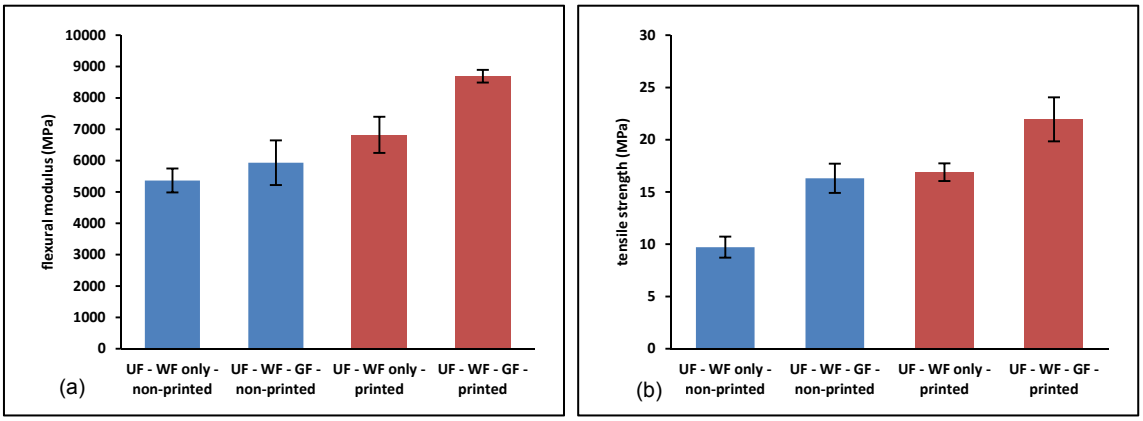

Fig. 3. Comparison of mechanical properties of printed and non-printed composites: (a) flexural modulus and (b) tensile strength. (UF = urea formaldehyde, WF = wood flour, GF = with glass fibres present).

compared to non-printed composites (12.3%). Fig. 4 shows SEM images of a cross-sectioned non-printed and printed sample (along the z-axis, build thickness). There are large voids observable within the wood cell walls compared to printed samples of the same

Table 3
Typical flexural and tensile properties for different wood and wood-based products [34].

Material		Flexural modulus (GPa)
Hardwoods	White oak	12.3
	Red maple	11.3
Softwoods	Douglas fir	13.4
	Western white pine	10.1
Panel products	Hardboard	3.1–5.5
	Medium density fibreboard	3.6
	Particleboard	2.8–4.1
	Oriented strandboard	4.4–6.3
	Plywood	7.0–8.6
Wood plastics		1.5–4.2
Material		**Tensile strength (MPa)**
Hardwood		73 (willow) – 121 (elm)
Softwood		59 (baldcypress) – 112 (larch)
Particleboard		6–11
Hardboard		23–38
Wood - plastics		6–25
Wood - cements		1–4

composition. Fig. 5 shows the extent of void inclusion in non-printed samples. The flexural stress-strain curves for samples with added glass fibres are shown in Fig. 2c and a comparison of the flexural properties of printed and non-printed samples is given in Fig. 3a. As with the non-glass fibre composites, the printed samples demonstrated a higher stiffness than the non-printed samples and had a higher flexural strength. As expected, they also had slightly better flexural properties than the urea formaldehyde-wood flour only printed samples, due to the presence of glass fibres. The flexural modulus of these composites was comparable to that of plywood (see Table 3).

Fig. 2d compares the tensile stress-strain curves of the composites containing glass fibres. Again, printed samples possessed a considerably higher tensile strength compared to non-printed samples due to lower void inclusion and directionality of glass fibres, acting parallel to tensile force. As expected, these composites also had a slightly higher tensile strength compared to the urea formaldehyde-wood flour only printed samples, due to the presence of glass fibres. The tensile strength of these printed samples was now comparable with that of hardboard (see Table 3).

As with the non-glass fibre printed composites, these improved mechanical properties could be due to a densification of the paste as it is extruded/deposited through the nozzle. Again, density measurements (Table 2) show that the cured printed samples have a higher density compared to cured non-printed samples, and a lower voids content (4.9%) compared to non-printed composites (12.3%).

The improved mechanical properties may also be due to glass fibre alignment during the extrusion process. Fig. 6 shows SEM images of printed and non-printed (moulded) cross-sections of composites of urea formaldehyde - wood flour with 10%wt. glass fibres. Fig. 6a and b clearly shows directional alignment of glass fibres along the scanning/printing direction. There appear to be many cracks within the sample compared to samples loaded without glass fibres; again this may be a result of the fibre inclusion itself. Fig. 6c and d show non-printed moulded samples; it can be clearly seen that there is no fibre alignment, thus contributing to a weaker structure compared to the printed sample when tested along the print direction. Fig. 7 shows sub-surface detail for (a) printed and (b) non-printed samples. Fig. 7a shows densely populated glass fibres directionally aligned within a surface crack. Fig. 7b shows highly randomised fibre alignment, with many fibres orientated perpendicular to the analysed surface. Adding glass fibres to the printed composition improved the tensile strength of samples by 30%; the highly orientated fibres improve the sample's resistance to deformation when positioned parallel to the force being exerted. As expected, the orientated fibres were therefore more effective in improving the tensile rather than the flexural

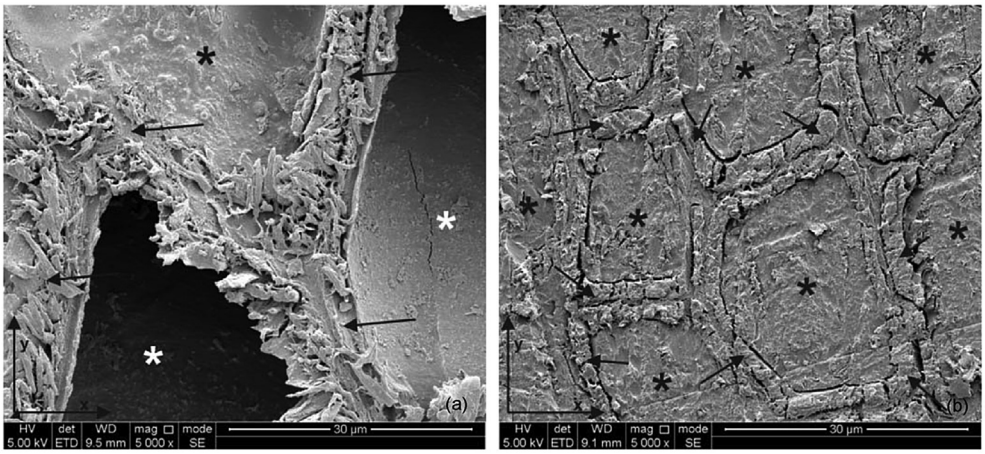

Fig. 4. Urea formaldehyde - wood flour; arrows indicate cell walls, black stars indicate UF, white stars indicate voids within the cell (a) non-printed samples (b) printed sample.

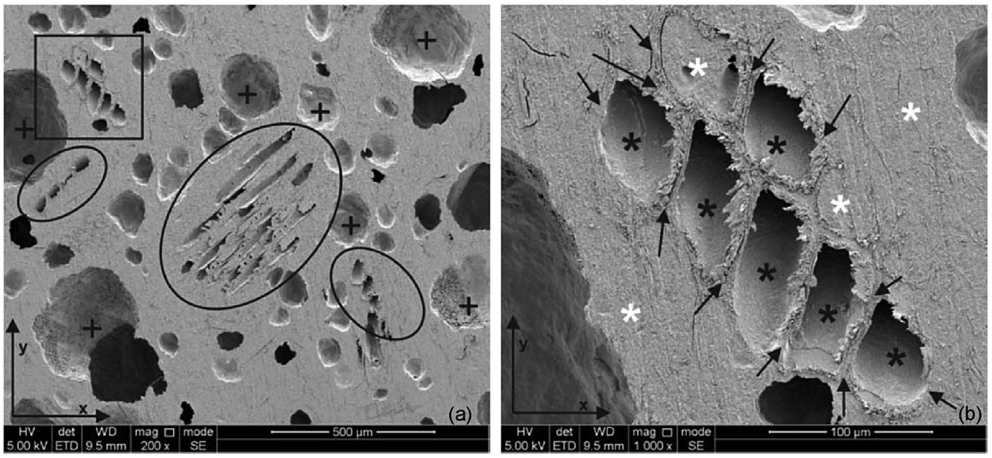

Fig. 5. Non-printed urea formaldehyde - wood flour samples: (a) wood particles indicated within circles in random orientation, + indicate gas pores. (b) Higher magnification image of box in Fig. 5a; arrows indicate wood cell wall, white stars indicate UF, black stars indicate voids within wood cell.

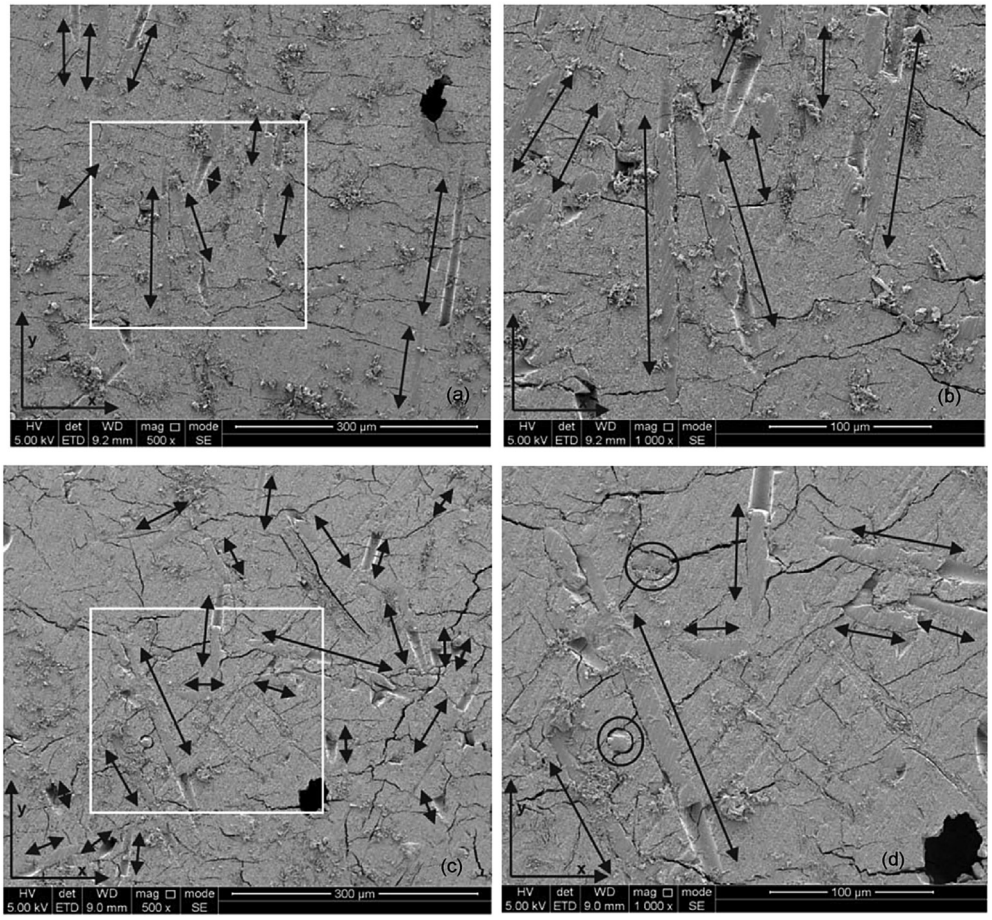

Fig. 6. Urea formaldehyde - wood flour with 10% wt glass fibres: (a) printed along the y-axis, evidence of directional alignment of fibres. (b) higher magnification image of box in Fig. 6a (c) moulded sample with randomly aligned glass fibres; small arrows indicate fibres that are almost perpendicular to analysed surface, longer arrows indicate fibres that are more parallel to analysed surface. (d) higher magnification of box in Fig. 6c moulded sample; circles indicate fibre running perpendicular to analysed surface.(a).

strength of printed samples. The significant difference between the flexural properties and tensile properties of the specimens occurs due to the multi-mode nature of the flexural test; combining tensile, compression and shear components in the results as opposed to the pure tensile test. As is typical of a polymer system such as the urea formaldehyde used in these experiments, compressive properties of this material are found to be higher than the tensile performance. Therefore, since the flexural test combines these properties, the compressive performance of the material dominates the flexural properties. The contribution of the compressive properties also accounts for the significant difference in flexural and tensile properties. Additionally, voids formed in the specimens

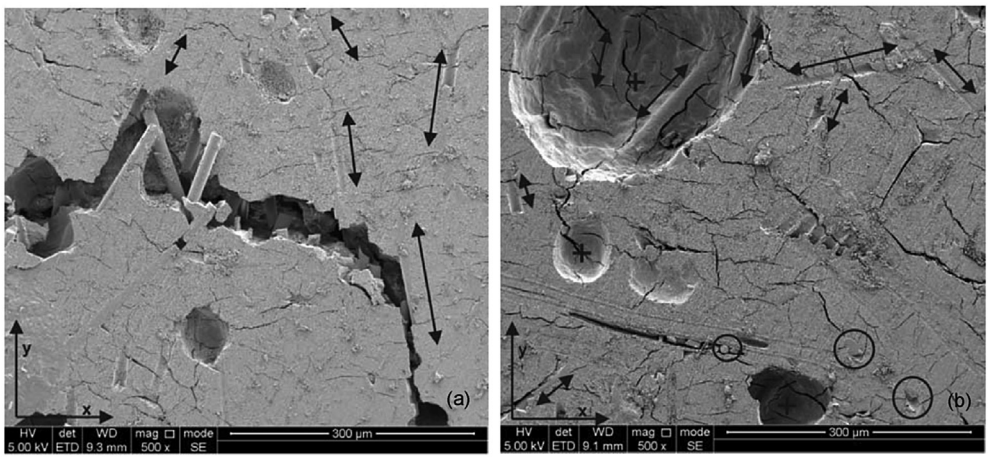

Fig. 7. Urea formaldehyde - wood flour with 10%wt glass fibres and sub-surface fibre detail: (a) printed along y-axis, densely populated and aligned fibres seen within a crack, some evidence of gas pores. (b) moulded sample with randomly aligned glass fibres, glass fibre detail shown within large gas pore.

provide a nucleation point for premature failure in the specimens. In the case of the tensile samples, voids present in the entire gauge length of the sample will additively contribute to specimen failure. In the case of the flexural samples, the maximum stresses are generated at the mid-section and outer surfaces, thus a much smaller area for voids to be present in the sample.

Although comparable in terms of mechanical properties to panel products, these printed products are not as mechanically strong as pure woods (Table 3). However, the highly porous structure of pure woods and the presence of hydroxyl groups in the cell wall, which attract moisture, make them liable to water adsorption/desorption leading to problems of dimensional stability [36]. Current solutions to this problem include chemical modifications such as cell wall bulking with acetic anhydride or polyethylene glycol, for example [36]. As with other composite panel products, problems with dimensional stability are less of an issue for wood - binder composites, consequently eliminating the need for expensive modifications. Here, binder resins react with the wood hydroxyl groups, thus making these reactive groups inaccessible. Furthermore, the high resin content significantly reduces the usually high void volume of wood, thereby removing the pathway for moisture diffusion in and out of the wood. This also reduces the chance of biological attack [37]. It should be noted that urea formaldehyde, used here as the binder for printing, is not suitable for products intended for outdoor use because prolonged moisture exposure leads to a breakdown of bond-forming reactions. However, there are other formaldehyde-based adhesives, such as phenol and melamine formaldehyde, which do not break down when exposed to moisture, and could be used as an alternative to produce printed products for outdoor applications [20].

4. Conclusions

In this study, wood – binder – glass fibre composites have been additively manufactured/3D printed using wood waste (wood flour) as the feedstock material and urea formaldehyde as the binder. Mechanical testing (flexural, tensile) has demonstrated that these products have improved mechanical properties compared to the non-printed samples manufactured in this study. Furthermore, these printed products have mechanical properties comparable to or even better than commercial panel products such as particleboard and fibreboard. The superior mechanical properties of the printed composites compared to non-printed composites are due to both a densification of the paste as it is extruded/deposited through the nozzle and fibre alignment induced by the printing process.

It is anticipated that the resolution of the deposited material and, therefore, its effect on the degree of densification and fibre alignment, will have a considerable effect on the mechanical properties of the product. Printing of more superior wooden products than those created in this study remains a possibility. The use of natural resins as adhesives such as lignin and tannin is also an interesting avenue of research, and there has been work into the manufacture of blends and part substitutions of formaldehyde resins with bioresins to provide environmentally friendly adhesives for use in the wood industry [38,39].

In regards to suitable applications for wooden products created by AM, it is believed that the small complex/bespoke components typically created by various AM techniques would not be of use to many engineering/industrial applications requiring a wooden composite. This material and process would have more value in the creation of on-site large bespoke products for the construction industry. Further to this, the products could be made lightweight by designing honeycomb voids within the structure, a feature difficult to achieve with conventional processing of wood. Furthermore, electronic devices such as sensors could be embedded during the layer wise manufacture and used to detect early signs of failure or interact with its environment. Though not completely sustainable due to the use of resins, this process and material has the potential to integrate an environmentally-friendly solution within the manufacturing industry with utilisation of a recycled and more sustainable feedstock.

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