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Recyclable hemp hurd fibre-reinforced PLA composites for 3D printing

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ABSTRACT

In this study, 3D printing filaments were produced from hemp hurd fibre-reinforced polylactide (PLA) composites. Hemp hurd microfibres were obtained through alkaline digestion followed by a bleaching treatment and were used to produce PLA-based composites with 20–40 wt% fibre content for fused deposition modelling. Tensile testing of 3D printed composites revealed a gradual increase of Young's modulus with the addition of fibres, reaching a maximum of 7.1 GPa for the 40 wt% composite - a two-fold increase to neat PLA. However, tensile strength was only improved for the 20 wt% formulation, with an increase of 8% in comparison with neat PLA. Nevertheless, the thermo-mechanical properties of the composites were significantly enhanced with the addition of fibres. In addition, physical objects were printed from the recycled filaments to assess their recyclability and printability. It was found that the recycled filaments maintained comparable mechanical properties and printability after three recycling cycles.

1. Introduction

The past decade has seen a notable increase in the interest in additive manufacturing (AM), otherwise called three-dimensional (3D) printing, for rapid fabrication of complex-shaped structures for various applications [1]. This is particularly based on the salient features of AM, such as structural and compositional gradation, high production efficiency, cost-effectiveness, and mathematically optimized designs [2,3]. Aligned with AM, there is a growing demand for the development of materials that support sustainability and a circular economy. This has led to increased attention to bio-derived materials, such as lignocellulose fibres (hemp, bamboo, flax, harakeke, wood) and biopolymers, such as polyhydroxyalkanoates (PHA) and polylactide (PLA) [4,5]. PLA, in particular, is widely used for fused deposition modelling (FDM) due to its renewability, biodegradability, good printability and mechanical properties [6]. However, the use of PLA is often limited by issues relating to its low stiffness and low thermo-mechanical stability. Different reinforcing materials, in special natural fibres, have been used to overcome these drawbacks [2,7–11]. Natural fibre-reinforced PLA composites provide a cost-effective, lightweight, recyclable, and environmentally sustainable approach to manufacturing products with varying levels of mechanical strength through conventional production techniques [6,7,9,12]. However, achieving high mechanical strength in natural fibre-reinforced PLA composites through FDM remains an ongoing challenge.

The main limitation of FDM for printed natural fibre-based polymer composites is the characteristic extrusion-related defects and poor interfacial bonding between the reinforcement and matrices [8]. In addition, it is generally difficult to eliminate inhomogeneities caused by the agglomeration of fibres. These limit the mechanical performance of composites, particularly when the fibre content is high. For example, Xiao et al. [11] reported that the tensile properties of 3D printed hemp hurd fibre reinforced PLA composites decreased significantly when the fibre loading exceeded 20 wt%. This was attributed to increased porosity and insufficient interfacial bonding. In another study, a significant amount of porosity (about 16%) was reported for a commercial filament composed of a PLA/poly hydroxyalkanoate (PHA) blend reinforced with 15 wt% wood fibre [10]. Similarly, poor interfacial bonding and high porosity were reported for 3D printed hemp/PLA composites containing 30 wt% fibre [13]. To overcome this challenge, different chemical, mechanical, and physical methods have been considered either to modify the fibre surface or the polymer matrix to improve the fibre-matrix interfacial bonding. Some of the notable efforts concerted towards improved physical and mechanical performance of PLA composites containing natural fibres are available in the literature [8,14,15]. It has been demonstrated that alkali-based treatments of natural fibres followed by a bleaching process improve fibre/matrix interfacial bonding and processability of the composites, which is highly beneficial for 3D printing applications [12,16]. Alkaline treatments remove amorphous components from the biomass such as hemicellulose and

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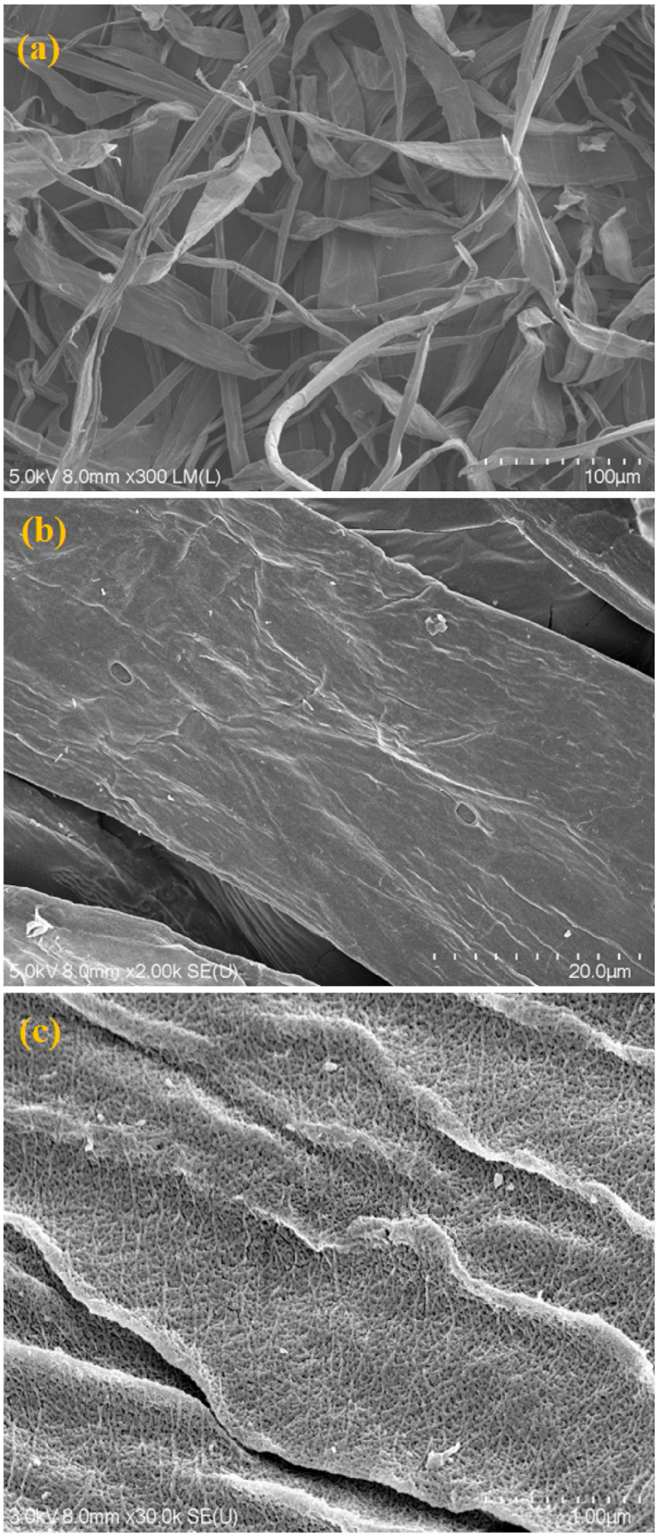

Fig. 1. SEM image of (a) hemp hurds, (b) single hemp hurd fibre, and (c) high magnification image of hemp hurd fibre surface.

lignin and separate/debond cellulose individual fibres. With bleaching, it is possible to further remove lignin from the fibre surface and expose more cellulose. By combining these two treatments, fibres with a better aspect ratio, surface roughness, and compatibility with the matrix can be obtained, resulting in composites with improved mechanical properties.

As detailed in the literature, different natural fibres have been compounded with PLA to develop composites. However, there is limited research on PLA composites reinforced with fibres extracted from hemp hurd. Hemp hurd is a byproduct of hemp fibre production and its primary use has been as a filler in the form of particles for polymers or used in the formulations of “hempcrete” [11,17–20]. In this work, hemp hurd was processed by alkaline and bleaching treatments and explored in its fibrous form. The fibres were incorporated into PLA to produce 3D printing filaments, which were further used to produce specimens for mechanical testing and physical objects. It is well known that waste, such as poorly mixed components, unused filaments, or defective printed objects, is generally generated in the 3D printing process. However, despite the increasing number of studies on 3D printing of PLA/natural fibre composites, there are not many considerations on the recyclability of the filaments or printed objects. Therefore, the filaments produced in this study were re-processed and reprinted several times to assess the effect of recycling on the mechanical properties of the composites. In addition, physical objects were printed from the recycled filaments, to determine the possible number of recycling steps it can withstand before a significant reduction in their printability.

2. Materials and methods

2.1. Materials

Polylactide (PLA) grade 2003D with melt flow index (MFI) of 6 g/10 min (210 °C, 2.16 kg) and specific gravity of 1.24 g cm^{-3} was purchased from NatureWorks®. The hemp stems were supplied by the Kokako Trust Ltd, NZ. Sodium hydroxide pellets, sodium sulphite powder, and hydrogen peroxide (30%) were procured from Sigma-Aldrich. Sodium silicate solution (acidimetric, Na_2O- 7.5–8.5 %, acidimetric, SiO_2 −25.5–28.5 % and density 1.35 g/ml) was supplied by Merck Millipore.

2.2. Fibre treatment

Air-dried hemp hurds were separated from hemp stems (1–1.5 m long) and then were cut into 2–3 cm lengths using a guillotine. Then, the hurds were weighed and subjected to digestion in a mixed solution of sodium hydroxide and sodium silicate following the procedure described previously [16]. After digestion, the hurd fibres were washed severally with water until the wastewater reached pH 7 and then the fibres were dried at 80 °C in an oven for 48 h. Digested fibres were further bleached using a solution of H_2O_2 and $NaSiO_3$, following the procedure described in the literature [16]. After bleaching, a Sunbeam Multigrinder with blunt blades running at a high rotational speed was used to defibrate the fibres.

2.3. Fibre Characterization

The diameter of the hemp hurds was obtained from images generated using an Olympus (model BX53) optical microscope, equipped with polarized light. About 100 fibres were analyzed as indicated on the representative image in Fig. S1 of the supplementary file, and the average fibre diameter was measured. In addition, the fibre surface was examined using a Hitachi S-4000 field emission scanning electron microscope, operated at 5 kV. The test material was placed on aluminium stubs, and sputter coated with palladium before observing it under the SEM.

2.4. Composite fabrication

Composites were first produced from PLA and hemp hurd fibres at different fibre content (20 wt%, 30 wt%, and 40 wt%). The matrix and fibre were compounded twice using a TSE-16-TC twin-screw extruder with a screw speed setting of 100 rpm at a temperature profile in the range of 165–175 °C. The extrudate was pelletized into 3 mm pellets using a Moretto GR knife mill and dried in a vacuum oven set at 60 °C for 2 h. Dried composite pellets were then fed into a single screw 3D

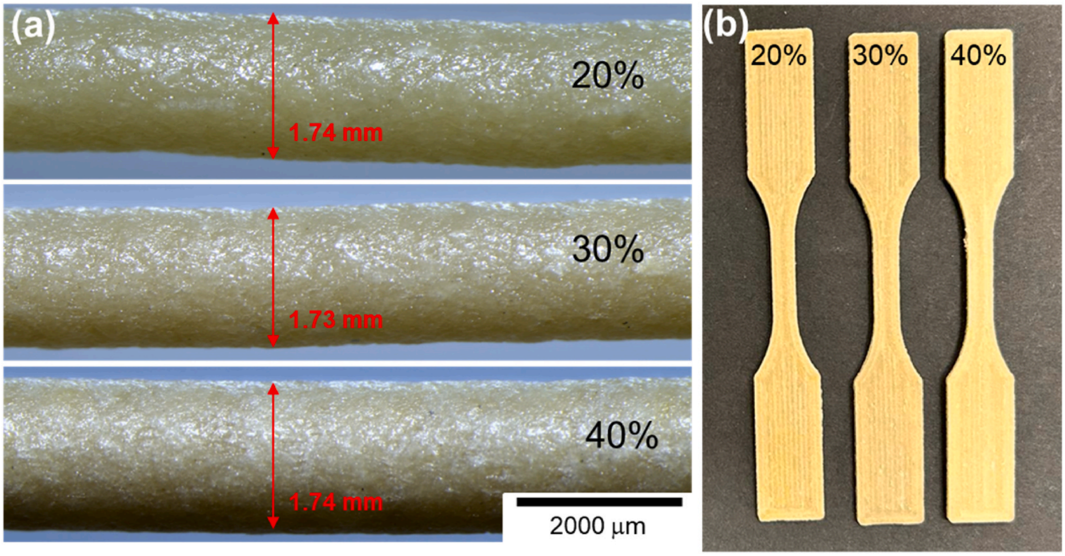

Fig. 2. Microscopic images of printing filaments (a) at different fibre content and digital images of their tensile testing specimens (b).

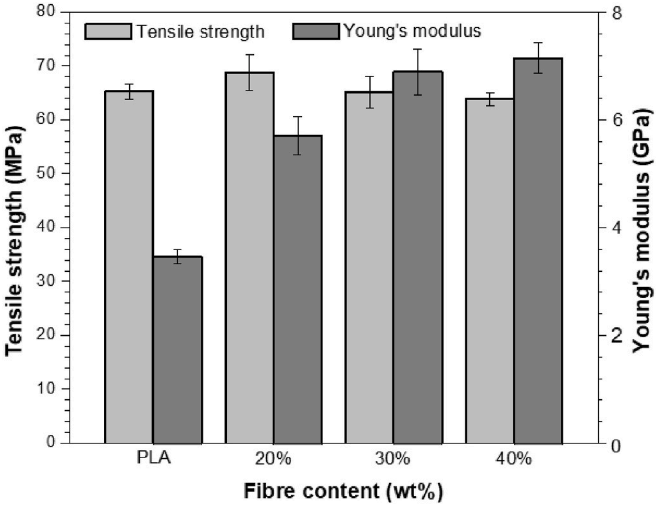

Fig. 3. Tensile strength and Young's modulus of 3D printed PLA, and PLA/hemp hurd fibre composites at different fibre content.

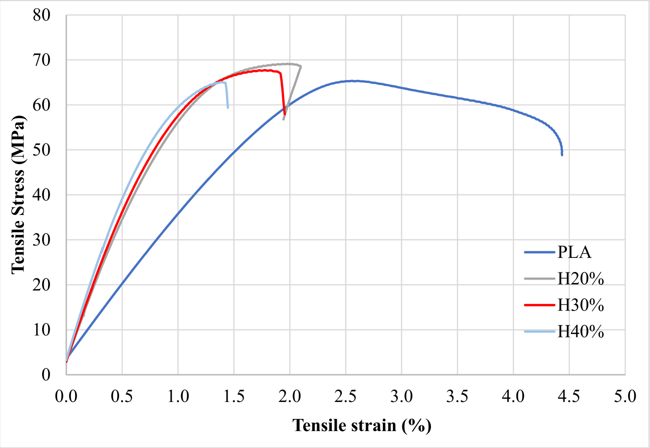

Fig. 4. Representative stress-strain curves of PLA and PLA/hemp hurd composites at different fibre content.

printing filament extruder (Filabot EX2 single screw extruder) at 180 °C to produce printing filaments. During filament production, the extrusion and spooling speeds were adjusted to produce filaments with a constant diameter of 1.75 ± 0.10 mm. Tensile testing samples were 3D printed according to ASTM D638 Type V specimens using a Maker Gear™ M2 desktop 3D printer. The 3D printer was equipped with a Simplify 3D® software package for slicing the computer-aided design (CAD) files and controlling the 3D printer. Samples were printed with a free-span nominal dimension of 3.18 mm, 1.50 mm, and 10.96 mm for width, thickness, and length, respectively. The printing filaments were vacuum dried for 2 h at 50 °C before printing, and samples were printed at a printing speed of 1800 mm/min, and a layer height of 0.1 mm using predetermined printing parameters. Briefly, the infill density was 100%, the nozzle diameter was 0.75 mm, the raster angle for all layers was 0^{o}, the bed temperature was 70 °C, and the nozzle temperature was 210 °C. Prior to tensile testing, the printed samples were kept in a conditioning chamber for 48 h at 23 °C and 50% relative humidity to ensure that all the samples are subjected to same environmental conditions before testing.

Based on the tensile strength and modulus recorded for the composites, and for ease of comparison, the 20 wt% hemp hurd fibre reinforced composite was selected for the recyclability studies. The printing filament produced from the selected composite category was recycled by pelletizing using a Moretto GR knife mill for the desired number of recycling steps (1, 2, 3, and 4). After each recycling step, new filaments were re-produced using the filament extruder and tensile testing samples were printed using the 3D printer following the procedure described above. A 3D printed object (vase) of the composite with 20 wt% fibre was also subjected to recycling in a similar manner as the filament. Samples printed from the selected composite category and its recycled filaments were given code names as R0, R1, R2, R3, and R4 to indicate the number of recycling steps where R0 is the virgin composite and R4 is the 4 times recycled composites.

2.5. Characterization of composites

The surface of the filaments produced by the single-screw extruder was observed using an Olympus stereo microscope (model SZX7). Tensile testing of the 3D printing filaments was performed by mounting the samples in specially designed 3D-printed tabs (described in Fig. S1 in the supplementary data). Tensile testing was conducted on an Instron® 5982 universal testing machine equipped with a 5 kN load cell. The test was run at a cross-head speed of 2 mm/min and a 10 mm extensometer

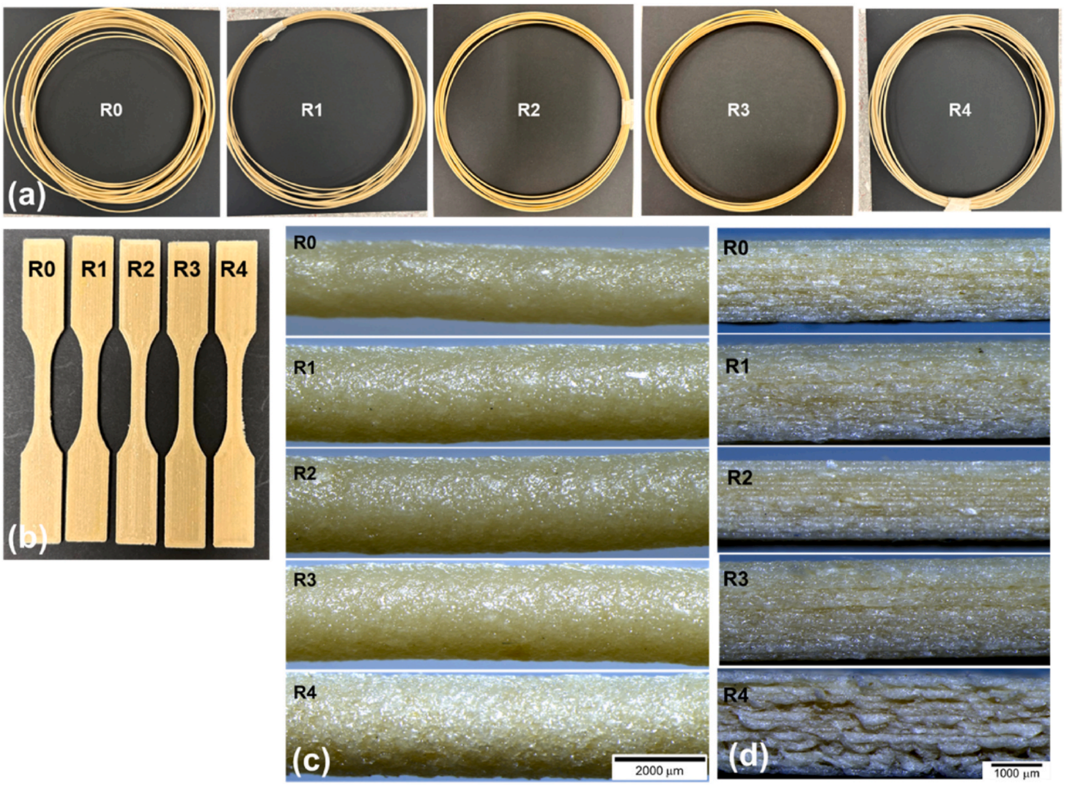

Fig. 5. Digital images of (a) extruded filaments, (b) tensile testing specimens, (c) stereo microscopy images of extruded filaments, and (d) side view of tensile testing specimens produced from PLA/hemp hurd fibre composite containing 20 wt% fibre, before and after each recycling steps.

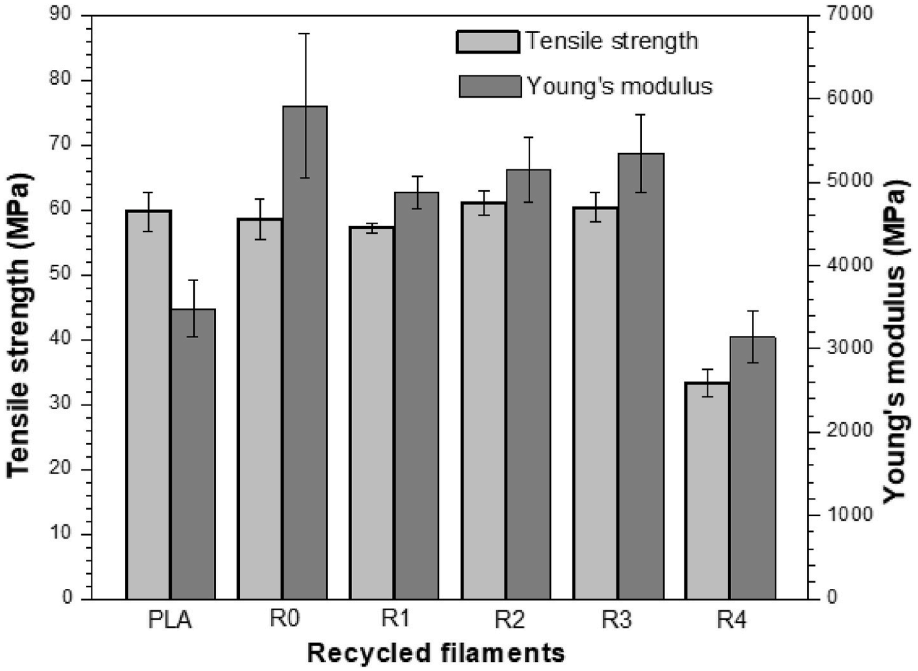

Fig. 6. Tensile properties of PLA, and PLA/hemp hurd fibre filaments (with 20 wt% of fibre content) before and after different number of recycling steps.

was used to measure the tensile strain. The ASTM D638 type V sample design was modified by increasing the length of the specimen (printing direction) by 15% to adequately accommodate the 10 mm extensometer. For each condition, five samples were tested, and the average values of tensile strength and Young's modulus were recorded. The tensile-tested fractured surface of the printed samples was analyzed through SEM using a Hitachi Regulus 8230 High-Resolution Scanning Electron Microscope at 3 kV, using a secondary electron detector. The samples were sputter coated with palladium in a Quorum Q150V plus before the analysis.

Thermogravimetric analysis (TGA) of the composites before and after recycling was performed using a PerkinElmer simultaneous

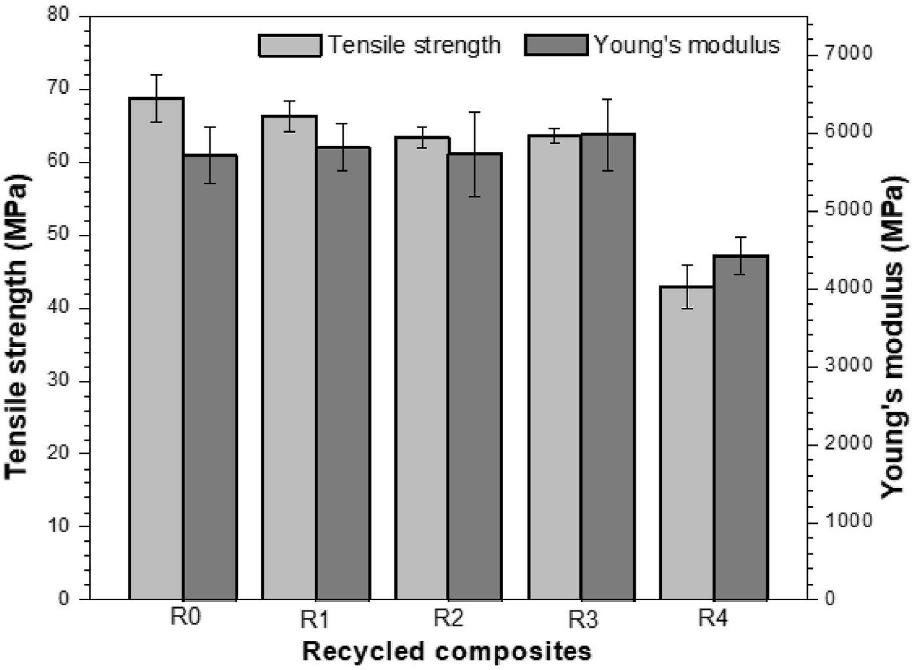

Fig. 7. Tensile properties of PLA/hemp hurd fibre 3D printed composites (with 20 wt% of fibre content) during different number of recycling steps.

thermal analyzer (STA 8000). The analysis was run at a rate of 10 °C/min over a heating range of 30 °C–600 °C under an argon flow of 40 mL/min.

Differential scanning calorimetry (DSC) of the 3D printed samples before and after recycling was conducted using a Netzch DSC3500 Differential Scanning Calorimeter. The samples were placed in aluminium crucibles and the instrument was run from 20 to 200 °C at 10 °C/min under a nitrogen flow of 60 mL/min. The glass transition (T_g), melting (T_m), and cold crystallization (T_{cc}) temperatures of the samples were obtained from the DSC thermograms.

The viscoelastic properties of the composites were assessed through dynamic mechanical analysis (DMA) using a Dynamic Mechanical Analyzer (PerkinElmer DMA800). The test was performed in a single cantilever mode and the samples were heated at 2 °C/min from 23 °C to 140 °C, at a displacement amplitude of 20 μm and a frequency of 1 Hz.

3. Results and discussion

3.1. *Morphological properties of fibre*

The SEM images of fibres extracted (digested and bleached) from hemp hurd are shown in Fig. 1. It can be seen from the SEM micrographs that the hurds appear as flat-shaped ribbon-like fibres (Fig. 1a). In addition, Fig. 1 (b) reveals the smooth appearance of the hurds. A closer view of the fibre shows that the fibre is composed of a network of microcellulose and has many pores (Fig. 1c) which could help to facilitate mechanical interlocking during composite production; this will be further discussed under the mechanical properties of the composites.

3.2. *Effects of fibre content on the properties of filaments and printed composites*

Fig. 2 shows the digital images of filaments produced from hemp hurd fibres and PLA at different fibre content (20 wt%. 30 wt%, and 40 wt%) (Fig. 2a), and the corresponding 3D printed tensile test specimens (Fig. 2b). It is worth mentioning that filaments were successfully produced at these fibre contents; however, with increasing fibre content, the surface of the filaments appeared to be rougher probably due to reduced wetting of fibres by the matrix. This shows a correlation between fibre content and filament surface roughness as reported previously in the literature [21].

Nevertheless, all the filaments were suitable for 3D printing. The mechanical properties of the printed neat PLA, and PLA/hemp hurd composites containing 20 wt%, 30 wt% and 40 wt% fibre content are presented in Fig. 3 while the representative stress-strain curves are illustrated in Fig. 4. As presented in Fig. 4, all the composites displayed brittle behaviours like PLA and the brittleness increased as the fibre content increased.

The brittleness of fibre-reinforced PLA composites generally increases as the fibre content increases, which is common with most fibre-reinforced composites. The incorporation of rubbery elastomers or plastics such as polycaprolactone, poly(butylene-adipate-*co*-terephthalate) (PBAT), and poly (butylene succinate) (PBS) may be used to reduce the brittleness of PLA [22–24]. This, however, can lead to issues relating to processability like in the case of PLA and PBS [25]. So, reactive processing with coupling agents can help to enhance processability, improve compatibility, and facilitate interfacial interactions in reinforced PLA composites. Nevertheless, it has been shown in the literature that the incorporation of rubbery additives often results in the sacrifice of the strength and Young's modulus of the composite. Therefore, in applications that require high mechanical strength, it is more desirable to utilize approaches that would support high mechanical performance such as treatment of fibres to facilitate good adhesion and interfacial interaction within the composite.

In this study, the composite with 20 wt% of fibres had the highest tensile strength (70 MPa), with an increase of 8% in comparison with neat PLA. However, the tensile strength of the composites decreased as the fibre content was increased from 20 wt% to 40 wt%. This is attributed to reduced wetting of the fibre by the matrix as confirmed by the increased roughness of the filament surface with higher fibre content (Fig. 2a). Composites with high fibre content also tend to have higher porosity which has a negative effect on the mechanical properties [11, 26]. In contrast, the tensile modulus of the composites increased with fibre content achieving a maximum of 7.1 GPa for the 40 wt% composite, which is 200% higher than neat PLA. The stiffness of hemp hurd fibres contributed to this positive outcome. Through these results, it is possible to observe the beneficial effects of using hemp hurd in its fibrous form instead of small particles. Xiao et al. [11] reported reduced

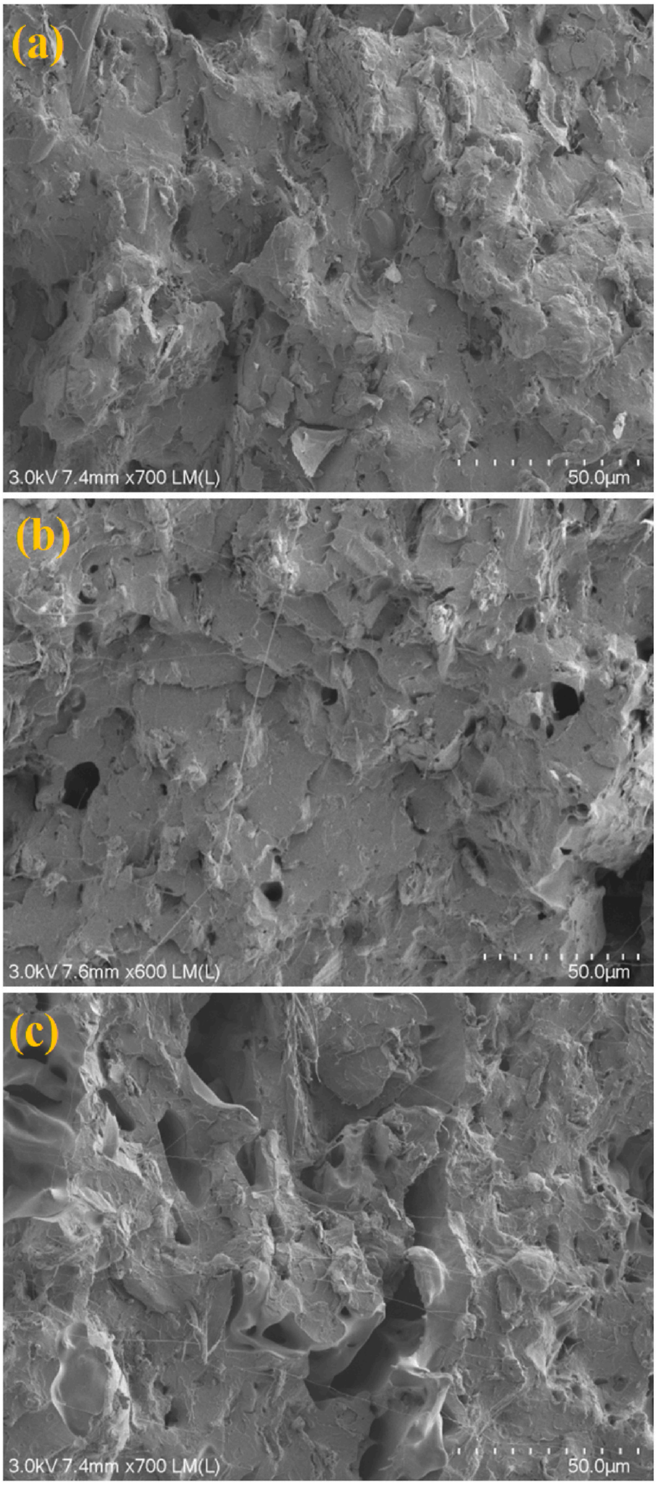

Fig. 8. SEM images of the fractured surface after tensile testing of composites printed using (a) non-recycled (R0), (b) one-time recycled (R1), and (c) four-times recycled (R4) hemp hurd/PLA filaments.

tensile and flexural strengths in FDM printed composites with contents of hemp hurd particles between 10 and 20 wt%. In that study, there were no significant changes in flexural modulus as well. The reduced mechanical properties in this case are justified by the increased porosity of the composites. By using hemp hurd fibres instead of particles it was possible to improve the reinforcement potential of the fibres and overcome the problems associated with increased porosity.

The major aim of this study was to investigate the recyclability of PLA/hemp hurd fibre filaments, and literature has shown that there is a possibility for higher porosity in filaments at high fibre content [27,28]. Therefore, based on the tensile properties of the composites and reports from similar studies in literature, the PLA/hemp hurds fibre composite containing 20 wt% fibre content was selected for further analysis and recyclability investigation.

3.3. Effects of recycling on the properties of filaments and printed composites

The digital images of filaments produced from PLA/hemp hurd fibre composite containing 20 wt% fibre and their printed tensile testing specimens before and after each re-processing step and side view of the printed tensile testing specimens are shown in Fig. 5. Based on Fig. 5a and b, it can be inferred that it was possible to reprocess the filaments up to 4 cycles, and test samples could be printed even after recycling four times (Fig. 5c). From the digital image, it is evident that there was no remarkable difference between the filament and printed sample surfaces, however, 4 times reprocessed filament surfaces were seen to have rough and uneven surfaces. This is believed to be due to the separation of the fibres from the matrix during the recycling steps, which can influence interlayer adhesion as evident from Fig. 5d where the side view of R4 is significantly different from other samples. In addition, a decrease in the molecular weight of PLA can also affect the rheology of the composite resulting in unstable flow of material during extrusion/printing.

The tensile strength and tensile modulus of the extruded filaments before and after recycling are presented in Fig. 6. The tensile strength and Young's modulus of the composite filament R0 is higher than that of neat PLA filaments which is due to the reinforcing effect of the fibres. Recycling up to 3 times did not significantly reduce the tensile strength and Young's modulus of the filaments. Beyond 3 times reprocessing, the tensile strength and Young's modulus of the filament (R4) dropped significantly. This effect could be due to reduced fibre length to below the critical fibre length (reducing the reinforcing potential of the fibres), increased porosity, separation of the fibre from the matrix, and decreased molecular weight of the polymer.

The tensile strength and tensile modulus of the composites printed from non-recycled and recycled PLA/hemp hurd filaments are presented in Fig. 7. As seen in the figure, the tensile strength and Young's modulus of the samples decrease as the number of recycling times increases. Up to three recycling steps, the drop in strength of the composites is minimal (~10%). The Young's modulus of the composites remains unchanged after being recycled three times. The tensile modulus of natural fibre reinforced polymer composites is often largely dependent on the modulus of the fibre, and the extent of interaction between the fibre and matrix [29]. Based on this, it can be inferred that the hemp hurd fibres were able to maintain their rigidity by resisting fracture and damage during recycling, thereby sustaining the tensile modulus of the composite. As seen in Fig. 7, beyond three recycling steps, there is a significant drop in the tensile strength and Young's modulus of the composite. This might be due to the reduction of fibre length below critical fibre length and separation of fibre from the matrix. This suggests that the recyclability of PLA/hemp hurd fibre composites is only possible over a given number of recycling steps (three times, based on this study), beyond this, the mechanical properties of the composite are excessively reduced.

The SEM images of the fractured surface after tensile testing of PLA/hemp hurd fibre composites are presented in Fig. 8. For ease of comparison, two representative images (R1 and R4) were compared with the unrecycled composite (R0). Fig. 8a shows a properly compounded composite with a small number of voids and fibre pull-out holes. In contrast, there are a significant number of voids and fibre pull-out sites on the fractured surface of the recycled composites (Fig. 8a and b), with more pores and voids evident in Fig. 8c (R4) which was subjected to four recycling steps. This is an indication of fibre separation from the matrix

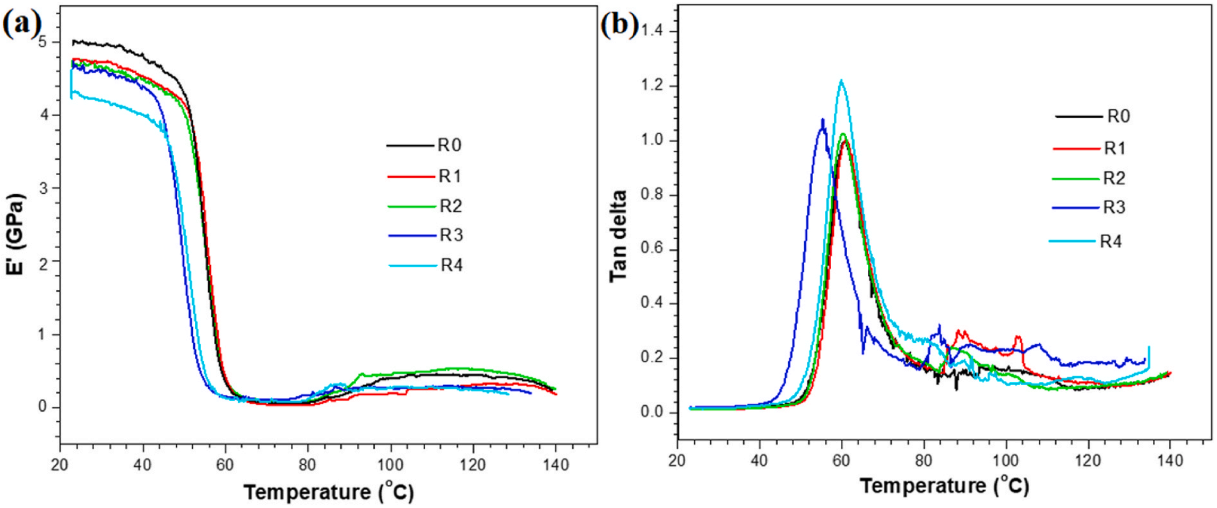

Fig. 9. (a) Storage modulus, and (b) tan delta curves of PLA/hemp hurd composites during different number of recycling steps.

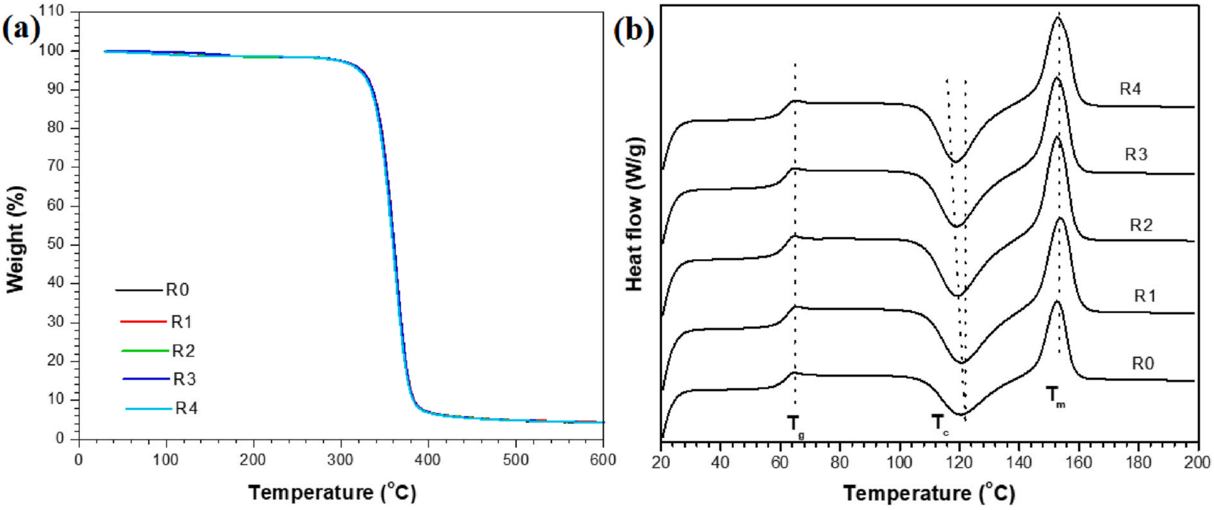

Fig. 10. TGA and DSC curves of PLA/hemp hurd filaments subjected to different numbers of recycling steps.

during recycling and might have contributed to the drop in tensile strength of the recycled composites as seen in Fig. 8 and discussed in previous paragraphs.

The effect of recycling on the viscoelastic properties of the PLA/hemp hurd fibre composites was further investigated through dynamic mechanical analysis. The storage modulus (E') curves of the PLA/hemp hurd fibre composites are illustrated in Fig. 9a. As presented in the figure, there is a gradual decrease in the E' of the composites as the number of recycling steps increases. The drop in E' became more significant when the number of recycling steps was increased from three to four and this aligns with the mechanical test result. It is well known that natural fibre reinforcements often offer increased stiffness to polymer matrices which in turn facilitates interfacial stress transfer within the composite [30]. In addition, the E' can be influenced by factors such as matrix type, filler type, filler distribution, and filler-matrix interfacial adhesion [30–32]. Therefore, it is believed that the drop in E' of the composites as the number of recycling steps increased is an indication of reduced interfacial bonding between PLA and the hemp hurd fibres. This would lead to fibre separation from the matrix and increased filament roughness as discussed in previous sections. Therefore, the effect of recycling on the interfacial interaction between PLA and hemp hurd fibre was further assessed through the damping parameter (tan delta (δ)).

The damping factor (tan δ) describes the relationship between the energy dissipated, and energy stored when a material is subjected to dynamic constraints. Generally, a higher tan δ is an indication of poor interfacial interaction, and vice versa. As seen in Fig. 9b, the tan δ peak increased as the number of recycling steps increased. This confirms that the interfacial relationship between PLA and the hemp hurd fibres decreased as the number of recycling steps increased, and this aligns with the mechanical test and microscopy results discussed in previous sections. In addition, a decrease is seen in the tan δ peak temperature in the samples R3 and R4, which indicates a decrease in the molecular weight of PLA.

The TGA curves are presented in Fig. 10a, while the DSC curves of the samples are illustrated in Fig. 10b. No significant changes in the thermogravimetric properties of the composites were seen even after recycling up to four times as seen in the TGA curves. This indicates that recycling did not result in thermal degradation of the hemp hurd reinforced PLA composites, which suggests that the materials can be used in applications that require good thermomechanical stability.

Similar to the TGA curves, there was no significant shift in the T_g, T_m, of the recycled composites, compared to the original composite. However, there is a little upward shift in the T_c of the recycled composites as seen in Fig. 10b. This slight shift in the Tc of the recycled composites can be attributed to the gradual pull out of fibre from the matrix as the

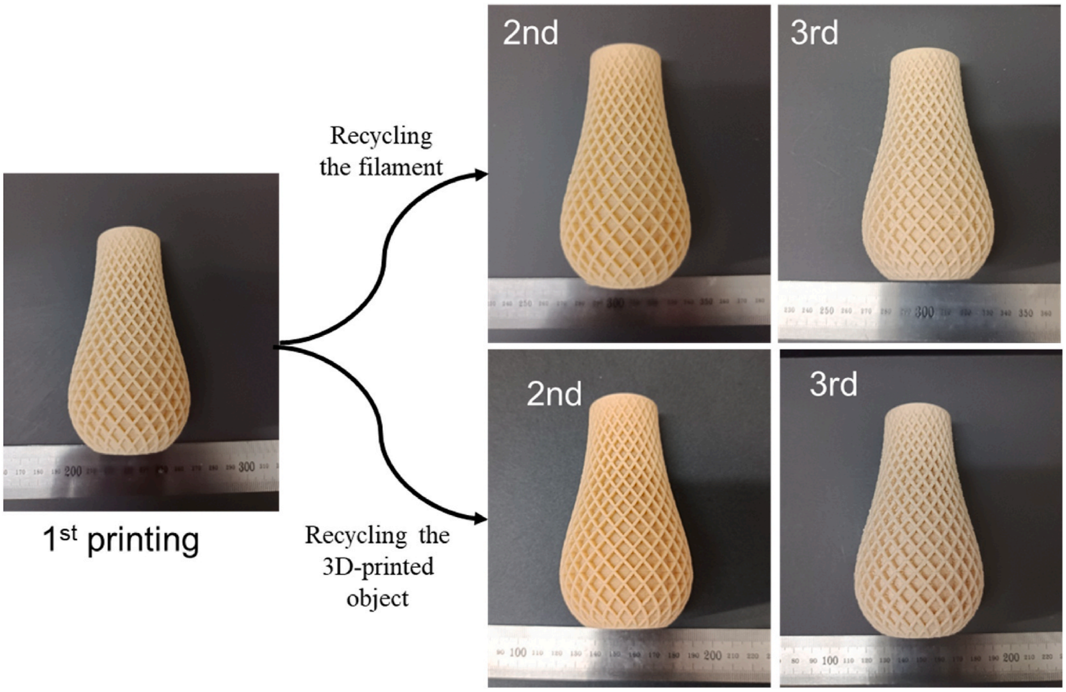

Fig. 11. Objects printed from hemp hurd/PLA filaments after different numbers of recycling.

number of recycling steps was increased.

Aside from the mechanical and viscoelastic properties of the printed tensile testing samples, physical objects were printed from the filaments, before and after recycling. The digital image of the vase printed from the filaments is shown in Fig. 11. As seen in the figure, objects can be reprinted from the filaments even after recycling up to three times. The filament subjected to four recycling steps could not be printed which suggests an undesirable drop in the printability of the filament after four recycling steps. The possible separation of fibre from the matrix during recycling might adversely affect the filament quality as seen in the tensile properties of filaments in Fig. 6. This is believed to have contributed to the significant drop in tensile properties of the four-time recycled composite (R4) as illustrated in Fig. 7. In addition, as seen in Fig. 5d, interlayer adhesion is affected by the extreme number of recycling steps. This is attributed to decreased interfacial bonding, perhaps due to fibre separation from the matrix during recycling. Xiao et al. [11] reported a direct relationship between poor interlayer adhesion, increased number of void formations, increased porosity, and decreased mechanical properties of 3D-printed natural fibre composites. Therefore, the drop in mechanical properties of the composites and the difficulty in printing at a higher number of recycling steps can be attributed to poor bonding.

4. Conclusions

Composites were produced from PLA and hemp hurd fibre. The reinforcing ability of hemp hurd fibres is portrayed in this study. The recyclability study showed that as the number of recycling steps increased, there was an increasing possibility for fibre separation from the matrix. Nevertheless, mechanical testing showed that PLA/hemp hurd fibre composites can be recycled for up to three times without catastrophic reduction in mechanical performance. In addition, thermal analysis confirms that the composites retained their thermal properties over the recycling steps which suggests that they can be explored for applications that require good thermal stability.

Declaration of Competing interest

Dear Editor, We are submitting the manuscript entitled “Recyclable hemp hurds fibre-reinforced/PLA composites for 3D printing” for consideration of publication in the Journal of Construction and Building Materials. We confirm that it has not been previously published or currently submitted elsewhere.

The authors confirm that no author has a conflict of interest with respect to the work reported or the submission of this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jmrt.2024.10.082.

References

[1] Bandyopadhyay A, Traxel KD, Koski C, Bose S. of Polymers. Additive Manufacturing 2019;25.
[2] Chacón J, Caminero M, Núñez P, García-Plaza E, García-Moreno I, Reverte J. Additive manufacturing of continuous fibre reinforced thermoplastic composites using fused deposition modelling: effect of process parameters on mechanical properties. Compos Sci Technol 2019;181:107688.
[3] Melenka GW, Cheung BK, Schofield JS, Dawson MR, Carey JP. Evaluation and prediction of the tensile properties of continuous fiber-reinforced 3D printed structures. Compos Struct 2016;153:866–75.
[4] Gauss C, Pickering KL, Muthe LP. The use of cellulose in bio-derived formulations for 3D/4D printing: a review. Composites Part C: Open Access 2021;4:100113.
[5] Mármol G, Gauss C, Fangueiro R. Potential of cellulose microfibers for PHA and PLA biopolymers reinforcement. Molecules 2020;25(20):4653.
[6] Akindoyo JO, Beg MDH, Ghazali S, Islam MR. Effects of poly (dimethyl siloxane) on the water absorption and natural degradation of poly (lactic acid)/oil-palm empty-fruit-bunch fiber biocomposites. J Appl Polym Sci 2015;132(45).

[7] Asyraf MRM, Syamsir A, Zahari NM, Supian ABM, Ishak MR, Sapuan SM, et al. Product development of natural fibre-composites for various applications: design for sustainability. Polymers 2022;14(5):920.

[8] Aydın M, Tozlu H, Kemaloglu S, Aytac A, Ozkoc G. Effects of alkali treatment on the properties of short flax fiber–poly(lactic acid) eco-composites. J Polym Environ 2011;19(1):11–7.

[9] Balla VK, Tadimeti JGD, Kate KH, Satyavolu J. 3D printing of modified soybean hull fiber/polymer composites. Mater Chem Phys 2020;254:123452.

[10] Le Duigou A, Castro M, Bevan R, Martin N. 3D printing of wood fibre biocomposites: from mechanical to actuation functionality. Mater Des 2016;96:106–14.

[11] Xiao X, Chevali VS, Song P, He D, Wang H. Polylactide/hemp hurd biocomposites as sustainable 3D printing feedstock. Compos Sci Technol 2019;184:107887.

[12] Akindoyo JO, Pickering K, Beg MD, Mucalo M. Combined digestion and bleaching of New Zealand flax/harakeke fibre and its effects on the mechanical, thermal, and dynamic mechanical properties of poly (lactic) acid matrix composites. Compos Appl Sci Manuf 2023;164:107326.

[13] Milosevic M, Stoof D, Pickering KL. Characterizing the mechanical properties of fused deposition modelling natural fiber recycled polypropylene composites. Journal of Composites Science 2017;1(1):7.

[14] Awad S, Siakeng R, Khalaf EM, Mahmoud MH, Fouad H, Jawaid M, et al. Evaluation of characterisation efficiency of natural fibre-reinforced polylactic acid biocomposites for 3D printing applications. Sustainable Materials and Technologies 2023;36:e00620.

[15] Ilyas RA, Zuhri MYM, Aisyah HA, Asyraf MRM, Hassan SA, Zainudin ES, et al. Natural fiber-reinforced polylactic acid, polylactic acid blends and their composites for advanced applications. Polymers 2022;14(1):202.

[16] Beg MDH, Pickering KL, Gauss C. The effects of alkaline digestion, bleaching and ultrasonication treatment of fibre on 3D printed harakeke fibre reinforced polylactic acid composites. Compos Appl Sci Manuf 2023;166:107384.

[17] Arehart JH, Nelson WS, Srubar IIIWV. On the theoretical carbon storage and carbon sequestration potential of hempcrete. J Clean Prod 2020;266:121846.

[18] Jubinville D, Chen G, Mekonnen TH. Simulated thermo-mechanical recycling of high-density polyethylene for the fabrication of hemp hurd plastic composites. Polym Degrad Stabil 2023;211:110342.

[19] Rhodes K, Pelaez-Samaniego MR, Kiziltas A, Preston J, Meysami M, Geda A, et al. Mechanical and viscoelastic properties of basalt-hemp hybrid reinforced polypropylene. J Thermoplast Compos Mater.0(0):08927057231177249.

[20] Stevulova N, Cigasova J, Purcz P, Schwarzova I, Kacik F, Geffert A. Water absorption behavior of hemp hurds composites. Materials 2015;8(5):2243–57.

[21] Tran TN, Bayer IS, Heredia-Guerrero JA, Frugone M, Lagomarsino M, Maggio F, et al. Cocoa shell waste biofilaments for 3D printing applications. Macromol Mater Eng 2017;302(11):1700219.

[22] Akindoyo JO, Pickering K, Beg MD, Mucalo M. Reactive compatibilization of harakeke fiber-reinforced poly (lactic) acid/polybutylene succinate blend. J Appl Polym Sci 2024:e56030.

[23] Fortelny I, Ujcic A, Fambri L, Slouf M. Phase structure, compatibility, and toughness of PLA/PCL blends: a review. Frontiers in Materials 2019;6:206.

[24] Mohammadi M, Heuzey M-C, Carreau PJ, Taguet A. Morphological and rheological properties of PLA, PBAT, and PLA/PBAT blend nanocomposites containing CNCs. Nanomaterials 2021;11(4):857.

[25] Supthanyakul R, Kaabbuathong N, Chirachanchai S. Random poly (butylene succinate-co-lactic acid) as a multi-functional additive for miscibility, toughness, and clarity of PLA/PBS blends. Polymer 2016;105:1–9.

[26] Muthe LP, Pickering K, Gauss C. A review of 3D/4D printing of poly-lactic acid composites with bio-derived reinforcements. Composites Part C: Open Access 2022;8:100271.

[27] Filgueira D, Holmen S, Melbø JK, Moldes D, Echtermeyer AT, Chinga-Carrasco G. 3D printable filaments made of biobased polyethylene biocomposites. Polymers 2018;10(3).

[28] Kariz M, Sernek M, Obućina M, Kuzman MK. Effect of wood content in FDM filament on properties of 3D printed parts. Mater Today Commun 2018;14:135–40.

[29] Beg MDH, Islam MR, Mamun AA, Heim HP, Feldmann M, Akindoyo JO. Characterization of polyamide 6.10 composites incorporated with microcrystalline cellulose fiber: effects of fiber loading and impact modifier. Adv Polym Technol 2018;37(8):3412–20.

[30] Saba N, Jawaid M, Alothman OY, Paridah MT. A review on dynamic mechanical properties of natural fibre reinforced polymer composites. Construct Build Mater 2016;106:149–59.

[31] Getme AS, Patel B. A review: bio-fiber's as reinforcement in composites of polylactic acid (PLA). Mater Today Proc 2020;26:2116–22.

[32] Rahman MZ. Mechanical and damping performances of flax fibre composites–A review. Composites Part C: Open Access 2021;4:100081.