Research Article

Investigation of Mechanical Properties of 3D Printed Biodegradable Polylactic Acid Reinforced with Paper Microcrystalline Cellulose

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Abstract
Fused Deposition Modeling (FDM) is an additive manufacturing technique that constructs objects layer by layer by depositing thermoplastic material through a nozzle. This method allows for the creation of intricate, custom designs that are often difficult to achieve with traditional manufacturing processes. To enhance the mechanical properties of composite materials, cellulose is used as a filler, which has shown significant potential in improving the physical and mechanical characteristics of polymer composites. In this study, waste paper is used to extract cellulose, resulting in microcrystalline cellulose (MCC), which is then used to reinforce the PLA matrix. Composite filaments containing different proportions of MCC (1%, 2%, and 3% by weight) are produced using a twin-screw extruder for subsequent 3D printing. The study examines the impact of MCC content on the structural, morphological, and thermal properties of the filaments and 3D-printed objects. Characterization methods include scanning electron microscopy (SEM), thermogravimetric analysis (TGA), and tensile tests. The results show that the addition of MCC does not cause chemical changes. For the 3D-printed samples, the tensile strength of neat PLA is significantly improved with the addition of 1% MCC and continues to increase with higher MCC concentrations.

Keywords: Eco-friendly fibers, Fused deposition modeling, Microcrystalline cellulose, PLA, Tensile strength

1 Introduction

The need for renewable alternatives to petroleum-based plastics is urgent due to the depletion of non-renewable resources and environmental concerns. Biodegradable materials, such as polylactic acid, have gained significant attention in recent years due to their potential applications in various industries including packaging, biomedical, and construction. However, polylactic acid (PLA) faces a challenge due to its limited strength, making it unsuitable for certain applications [1].

3D printing of polymer composites has the potential to address the limitations of pure polymer products, with promising results in developing new printable composite materials reinforced by particles, fibers, or nanomaterials [2]. Ning *et al.*, noted that adding carbon fiber to plastic materials can increase tensile strength and Young's modulus but may decrease toughness, yield strength, and ductility because excessive carbon fiber content can lead to increased porosity and decreased mechanical properties [3]. Filaments with embedded short carbon microfibres showed better print capabilities and were suitable for use with standard printing methods [4].

Similarly, Tao *et al.*, found that the WF/PLA composite filament was found to be suitable for 3D printing using the FDM process. Also, the addition of wood flour to the PLA matrix changed the microstructure and enhanced the initial deformation resistance of the composite, while also affecting the thermal degradation temperature [5].

Wang *et al.*, introduced nanoparticle impregnation modification for producing natural fiber reinforced composite materials with strong interfacial adhesion for 3D printing. Nano $CaCO_3$ impregnation significantly improved tensile strength compared to MCC and CNF impregnation; the reinforcement effect decreased above the glass transition temperature (Tg) [6]. Mathew *et al.*, [7] and Murphy *et al.*, [8] explored the addition of microcrystalline cellulose (MCC) and surface-modified cellulose to PLA, respectively. While Mathew *et al.*, observed an increase in storage modulus but decreased crystallinity and mechanical properties, Murphy *et al.*, demonstrated enhanced crystallinity and improved storage modulus through surface modification.

Wang *et al.*, studied the thermal behaviour of nanocellulose-filled filaments for 3D printing. They found that the addition of nanocellulose improves the thermal stability of PLA filament, with 1 wt % addition of nanocellulose resulting in the highest thermal stability [9]. Noesanto *et al.*, found the presence of cellulose nanocrystals (CNCs) in the PLA composite filaments improved crystallinity and thermal stability, enhanced tensile strength, and increased water absorption. The optimal improvement in tensile strength was observed at 1 wt% CNCs. The 3D printed samples containing CNCs showed lower water absorption than the neat PLA [10].

The strength of the 3D printed PLA, suitable for use in biomedical applications, has shown significant weight reduction over a period of 30 to 60 days in landfill tests. Moreover, the 3D printed PLA has good strength compared to ABS, indicating its potential as an alternative material, and the fabrication process is eco-friendly, with positive signs of degradation under standard atmospheric conditions [11], [12]. Cellulose nanofibers derived from waste paper exhibit promising potential for enhancing the mechanical and physical characteristics of polymer composites, especially in the context of food packaging applications [13].

Meanwhile, studies have revealed that varying FDM process parameters influence the mechanical properties of printed objects. Increasing printing layer thickness leads to higher water absorption and lower mechanical properties while decreasing printing layer thickness results in improved tensile and bending properties due to higher material density in the same volume [14]–[16]. Additionally, the printing temperature has an impact on the transparency of 3D-printed specimens [17].

Moreover, challenges in continuous fiber printing have led to void formation during printing and poor adhesion of fibers and polymer matrix. On the other hand, the introduction of fillers leads to blockage, wear, non-adhesion, and increased curing time. Hence, there is a need for further research on modeling and simulating the structures produced by the additive manufacturing process [18]–[20]. These studies provide valuable insights into how combining cellulose-based materials with polymers in 3D printing improves interactions and enhances mechanical properties, advancing additive manufacturing.

In this research, we explore creating eco-friendly, strong, and versatile materials by synthesizing microcrystalline cellulose-reinforced PLA filaments for 3D printing. Our study directly addresses the modern manufacturing challenge of needing sustainable, high-performance materials that work seamlessly with 3D printing technologies.

2 Materials and Methods

Material Selection: Matrix: Polylactic Acid (PLA)
Reinforcement: Microcrystalline Cellulose (MCC) from waste paper.

2.1 *Extraction of paper micro cellulose from waste paper:*

The waste paper utilized in this study, primarily sourced from used and printed materials, undergoes a meticulous preparation process to ensure optimal conditions for microcrystalline cellulose (MCC) extraction. Initial steps involve cutting the waste paper into smaller pieces to facilitate subsequent processing. To eliminate contaminants and impurities thoroughly, the paper is then cleaned in hot water.

Subsequently, a 2% NaOH treatment is applied for 3 h at 80 °C with stirring, followed by immersion in a solution containing 10% NaOH and 15% H_2O_2 for 3 h at room temperature. The bleached sample undergoes multiple rinses until reaching a neutral pH.

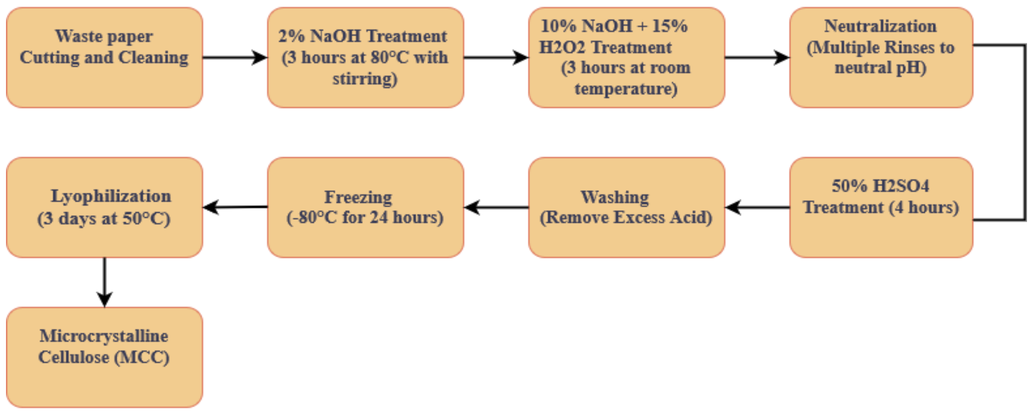

Figure 1: Processing of Microcrystalline Cellulose (MCC) from waste paper.

Further treatment with 50% H_2SO_4 for 4 h, followed by extensive washing, ensures the removal of excess acid, resulting in a refined paper product. The final steps involve freezing the sample in a deep freezer for 24 h at –80 °C and lyophilizing it for 3 days at 50ºC to obtain the micro cellulose powder.

Figure 1 shows a graphical representation depicting the processing of Microcrystalline Cellulose (MCC) from waste paper. This detailed process chain is crucial for obtaining high-quality MCC and contributes to the research's emphasis on sustainable and efficient material preparation.

2.2 *Brabender mixer:*

The preparation of the PLA/MCC composite filaments commenced with the meticulous drying of PLA pellets and MCC powder in an oven at 90 °C for 3 h. This step aimed to eliminate any adsorbed moisture, ensuring the integrity of the ensuing extrusion process. Subsequently, a Brabender mixer was employed for the amalgamation of PLA pellets and MCC powder at varying weight percentages—1%, 2%, and 3%. The mixing process unfolded at a temperature of 195 °C, carefully chosen based on our preliminary research.

The resulting composite filaments, namely PLA-1 MCC (1wt%), PLA-2MCC (2wt%), and PLA-3MCC (3wt%), were fabricated using an extruder, operating at a constant speed and temperature of 195°C. This controlled environment guaranteed the reproducibility of the filaments and their distinct MCC content. Notably, a neat PLA/MCC composite, devoid of additional cellulose, was also produced to serve as a control group in subsequent analyses.

The composited mixture, observed to be hard and lumpy, posed challenges for the 3D printing process due to its unwieldy size. The significance of this step lies in determining the printability and structural integrity of the composite filaments, shedding light on their applicability in real-world scenarios.

2.3 *Twin screw extruder:*

Following the Brabender mixer phase, the dried PLA pellets and MCC powder underwent a similar treatment for MCC content at 1%, 2%, and 3% in the twin screw extruder. This process, conducted at a temperature of 200 °C, aimed to achieve optimal blending and distribution of MCC within the PLA matrix. The resulting filaments, denoted as PLA-1MCC, PLA-2MCC, and PLA-3MCC, were then carefully cooled and collected for subsequent characterization and tensile testing. Figure 2 shows the prepared samples.

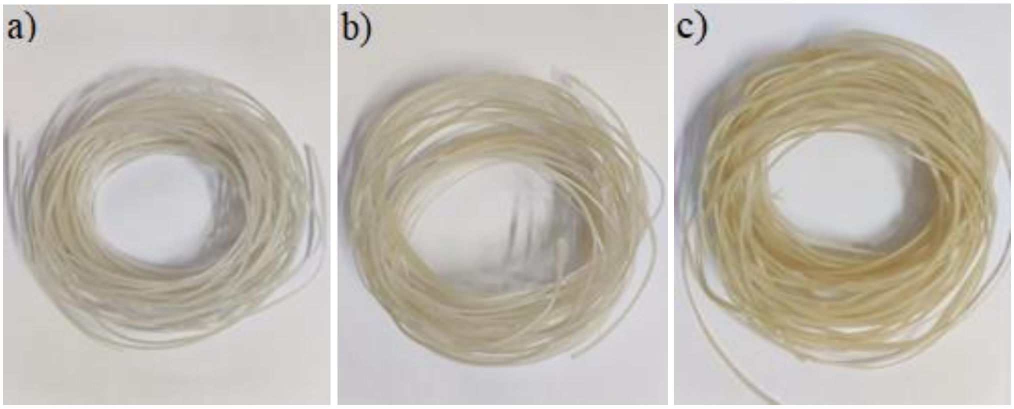

Figure 2: From left to right - the various filaments obtained from the extruder containing varying compositions of filament (a) PLA + microcellulose (1%) (b) PLA + microcellulose (2%) (c) PLA+ microcellulose (3%).

This step in the fabrication process is crucial for examining the impact of different blending techniques on the mechanical properties of the composite filaments. The choice of extrusion temperature is particularly critical, as it influences the homogeneity of the mixture and, consequently, the performance of the printed structures.

2.4 *3D printing of filament:*

Armed with the fabricated PLA/MCC composite filaments, the next phase involved their application in 3D printing. The samples as shown in Figure 3, designed in accordance with ASTM D638-10 type V specifications (63.50 × 9.53 × 3 mm^3), were printed using an FDM 3D printer (Ultimaker 2+). The printing parameters include a 0.4 mm nozzle size, 200 °C nozzle temperature, 60 °C bed temperature, 50% infill degree, 90° orientation angle, 0.3 mm layer thickness, and a print speed of 45 mm/s.

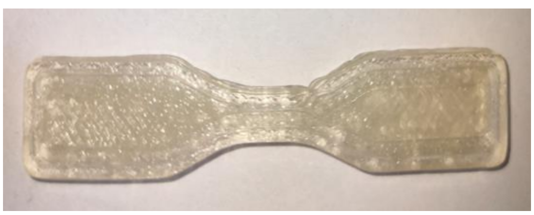

Figure 3: 3D printed PLA/MCC composite structure.

These parameters were optimized to strike a balance between printing efficiency and the mechanical integrity of the resulting structures. This comprehensive approach to material preparation ensures a thorough understanding of the interactions between PLA, MCC, and the 3D printing process, setting the stage for in-depth analyses in subsequent sections.

2.5 *Testing*

In our study, we conducted tensile testing to explore the impact of microcrystalline cellulose (MCC) on the mechanical properties of Polylactic Acid (PLA) and PLA/CNF composites for that, we used the Universal Testing Machine (UTM) to determine tensile strength, focusing on crucial properties like ultimate tensile strength and breaking strength. This allowed us to understand how the material responds under tension. Moving on to microstructural analysis, the Scanning Electron Microscope (SEM) revealed the intricate topography and composition of the samples. For a deeper exploration of thermal behavior, we employed Thermogravimetric Analysis (TGA). This technique involved subjecting samples to controlled temperature programs while measuring weight changes. TGA uncovered details about thermal stability, decomposition kinetics, and other crucial thermal properties.

3 Results and Discussion

3.1 *Tensile testing*

The tensile testing of 3D-printed PLA and PLA/Cellulose Nanofiber (CNF) composites was conducted to assess the impact of microcrystalline cellulose (MCC) reinforcement. The specimens were designed with specific dimensions (average thickness of 3.8 mm, width of 3.95 mm, and a gauge length of 25 mm) to ensure accurate measurement of material behavior under tension.

Neat PLA: The tensile test for neat PLA revealed a maximum force of 434.263 N and a maximum stress of 33.1356 N/mm². These values serve as a baseline to compare the mechanical properties of the PLA composites.

PLA with 1% MCC: The introduction of 1% MCC to PLA resulted in an increased maximum force of 568.351 N and a maximum stress of 37.526 N/mm². This improvement indicates enhanced material strength and elongation at break compared to neat PLA.

PLA with 2% MCC: For the composite with 2% MCC, the maximum stress rose to 42.895 N/mm², and the maximum force reached 672.275 N, further demonstrating the beneficial impact of MCC on tensile strength.

PLA with 3% MCC: The composite with 3% MCC content showed the most significant enhancement, with a maximum stress of 53.1411 N/mm² and a maximum force of 820.286 N.

Table 1 in the study provides a comprehensive summary of these results, detailing the variations in maximum force, stress, and elongation for each sample. The table effectively illustrates the progressive improvement in mechanical properties with the increasing addition of MCC, highlighting the potential of MCC as a reinforcing agent in PLA composites for 3D printing applications [21], [22].

Table 1: Tensile properties of PLA and PLA/MCC composites.

Materials	Maximum Force (N)	Maximum Stress (N/mm²)	Max. Stroke (%)	Break Stress (N/mm²)	Break Stroke (%)	Break Force (N)
PLA	434.263	33.1536	2.80707	24.4707	3.857	320.522
PLA + 1% MCC	568.351	37.526	3.01254	32.5812	4.223	420.526
PLA + 2% MCC	672.275	42.895	3.4684	37.8562	4.796	554.627
PLA + 3% MCC	820.286	53.1411	3.7604	45.9077	5.1304	709.904

3.2 *SEM analysis*

Scanning electron microscopy (SEM) was employed to examine the morphology of the tensile fractured surface of 3D printed PLA/MCC composites.

The SEM analysis revealed distinct characteristics for neat PLA compared to PLA/MCC composites. Neat PLA exhibited a relatively smooth fracture surface indicative of brittle fracture as evident from Figure 4(a) [23]. In contrast, the PLA/MCC composites showed a relatively flat fracture surface characteristic of brittle fracture with slight plastic deformation in the polymer matrix and no necking in any of the filaments.

The SEM images (Figure 4(b)–(d)) indicated a homogeneous distribution of MCC within the PLA matrix across all samples, as no clusters or large aggregates of MCC were observed. This uniform distribution suggests an enhanced interaction between MCC and PLA, which is essential for improving the mechanical properties of the composite.

The presence of voids in the fracture surface was attributed to the extrusion process and poor interfacial adhesion between the reinforcement materials and the matrix, which affects the mechanical properties of the extruded filaments [22].

Figure 4(b) illustrates the SEM image of the surface of the composite filament containing 1 wt% whereas, Figure 4(c) illustrates the SEM image of the surface of the composite filament containing 2 wt%. The SEM image of the fracture surface of the filament containing 3 wt% MCC is shown in Figure 4(d). All PLA/MCC composite filaments generally exhibited a relatively flat fracture surface, indicative of brittle fracture, with slight plastic deformation in the polymer matrix and no necking is observed. This correlates with the relatively low elongation at break value (2–5%) discussed in the tensile test section [22].

A common feature observed in the fracture surfaces of all extruded PLA/MCC composite filaments was the presence of voids, as seen in Figure 4(b)–(d). The formation of these voids can be attributed to two primary factors. First, voids were generated during the extrusion process when the melted filament was extruded from the nozzle. These voids then enlarged as the melt exited the die due to the pressure differential between the inside and outside of the die [22]. Second, poor interfacial adhesion between the reinforcement materials and the PLA matrix contributed to void formation. The presence of these voids adversely affects the mechanical properties of the extruded filaments [22].

The SEM image analysis underscores the importance of ensuring a uniform distribution of MCC and enhancing the interfacial adhesion between the matrix and reinforcement materials [22]. These factors are critical for improving the overall mechanical performance of PLA/MCC composites.

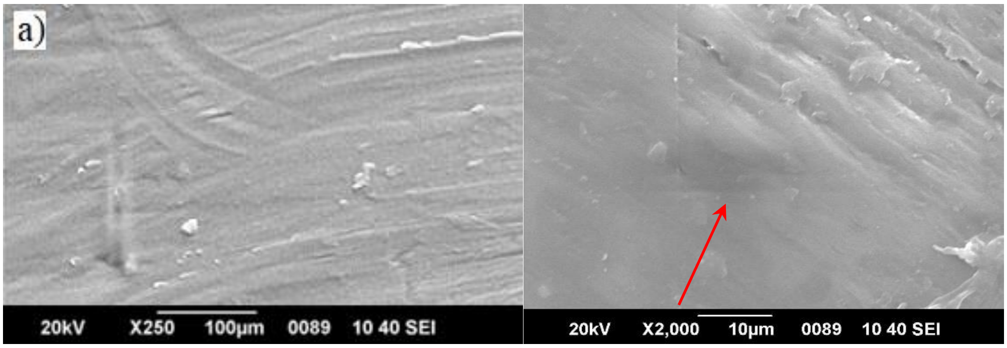

Figure 4(a): SEM fractography for Neat PLA.

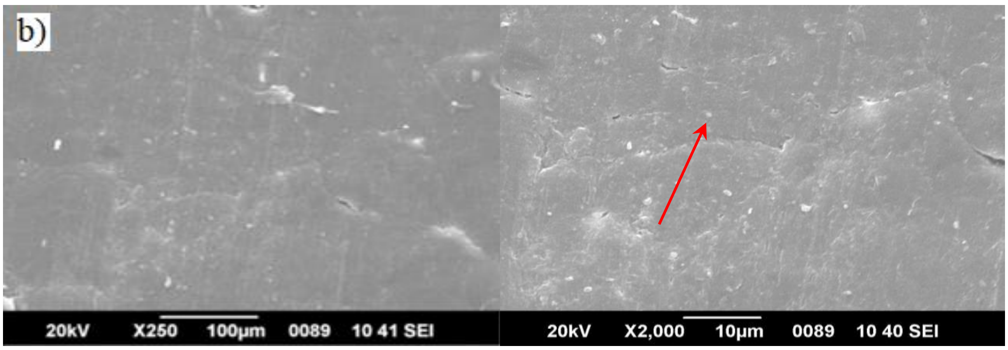

Figure 4(b): PLA with 1% MCC content.

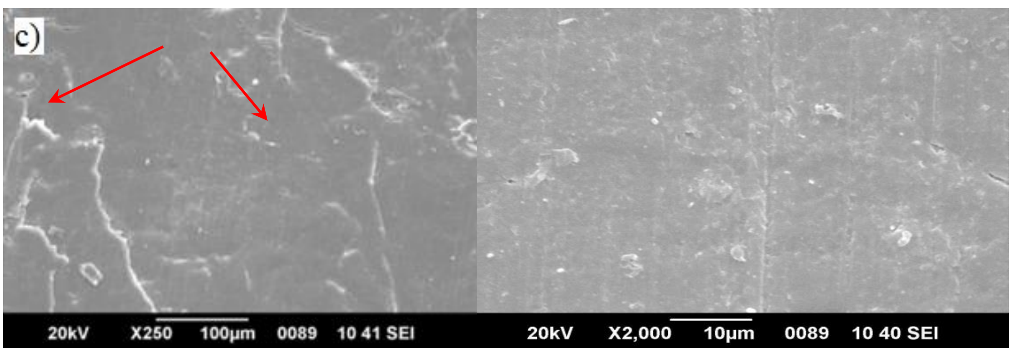

Figure 4(c): PLA with 2%MCC content.

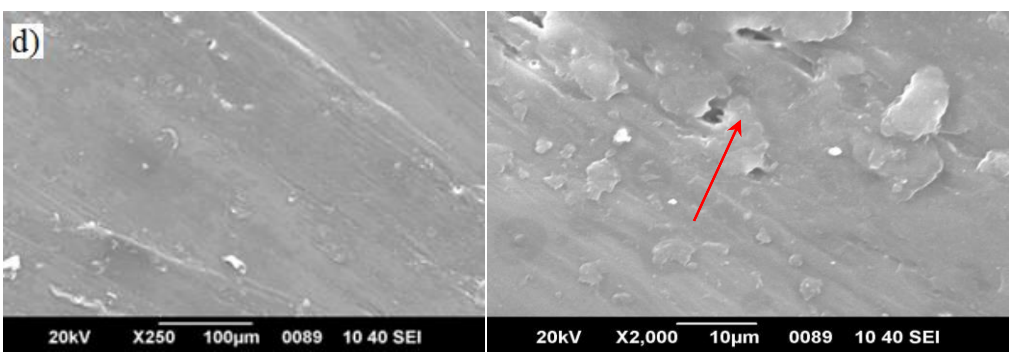

Figure 4(d): PLA with 3% MCC content.

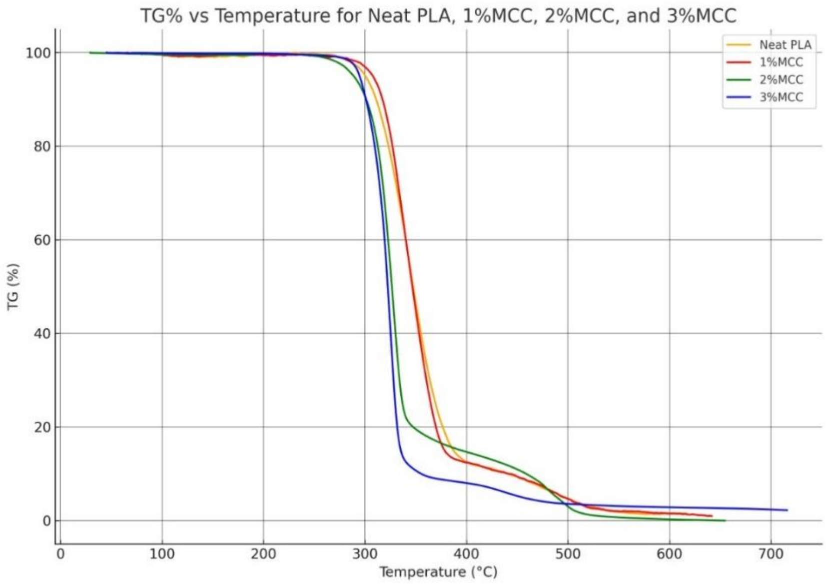

Figure 5: Thermogravimetric analysis of PLA/MCC composite material samples.

4.3 *Thermogravimetric analysis (TGA)*

Thermograms from Simultaneous Thermal Analysis (STA), which combines Differential Scanning Calorimetry (DSC) and Thermogravimetric Analysis (TGA) are used to analyze the thermal properties and stability of composites [21]. The thermal stability analysis of PLA-MCC composite samples with 1%, 2% and 3% MCC content was done against pure PLA.

From Figure 5 it is clear that pure PLA typically melts between 115–132 °C and starts to break down at around 265 °C, with the most rapid decomposition occurring near 336.84 °C which is in agreement with the previously published results [24]–[26]. PLA with 1% MCC exhibited the best thermal stability as the material could withstand higher temperatures before melting at 352.3 °C and starting an exothermic reaction at 438.2 °C while PLA with 2% and 3% MCC demonstrated lower temperature resistance, with a rapid decomposition occurring between 310 °C and 330 °C.

The data shows that when compared to pure PLA the MCC composites are less stable at high temperatures with an increase in weight percentage of MCC. In summary, adding MCC to PLA makes it less stable at high temperatures, with the 1% MCC composite being the most stable among the ones tested.

4 Conclusions

This research demonstrates that adding microcrystalline cellulose (MCC) from waste paper to PLA composites enhances their mechanical properties. In FDM 3D printing, MCC improves the structural, morphological, and thermal properties of PLA, offering a sustainable solution for various industrial applications. Including 1%, 2%, and 3% MCC by weight significantly increases PLA's tensile strength, with higher MCC content resulting in greater force and stress. PLA/MCC composites exhibit similar thermal stability to neat PLA at low MCC percentages, with higher melting points and exothermic reaction temperatures, especially at 1% MCC.

SEM analysis shows a homogeneous distribution of MCC in the PLA matrix, indicating strong interaction and uniformity, crucial for improved mechanical properties. Using waste paper for MCC supports environmental sustainability, and PLA/MCC composites are biodegradable, making them suitable for eco-friendly applications. The improved properties and biodegradability of PLA/MCC composites make them ideal for industries such as packaging, biomedical, and construction. These findings encourage further exploration of PLA/MCC composites for sustainable, high-performance materials in additive manufacturing.

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Author Contributions

All authors have read and agreed to the published version of the manuscript. The individual contributions can be summarised as follows: T.A.: conceptualization, investigation, methodology, writing original draft; R.C.R.: investigation, writing original draft, reviewing and editing; A.A.: research design, writing—reviewing and editing; E.J.: research design, writing—reviewing and editing; V.D.N.: research design, writing—reviewing and editing;

Conflicts of Interest

The authors declare no conflict of interest.

References

[1] M. Bardot and M. D. Schulz, "Biodegradable poly (Lactic acid) nanocomposites for fused deposition modeling 3D printing," *Nanomaterials*, vol. 10, no. 12, pp. 2567, 2020, doi: 10.3390/ nano10122567.

[2] X. Wang, M. Jiang, Z. Zhou, J. Gou, and D. Hui, "3D printing of polymer matrix composites: A review and prospective," *Composites Part B: Engineering*, vol. 110, pp. 442–458, 2017, doi: 10.1016/j.compositesb.2016.11.034.

[3] F. Ning, W. Cong, J. Qiu, J. Wei, and S. Wang, "Additive manufacturing of carbon fiber reinforced thermoplastic composites using fused deposition modelling," *Composites Part B: Engineering*, vol. 80, pp. 369–378, 2015, doi: 10.1016/j.compositesb.2015.06.013.

[4] L. G. Blok, M. L. Longana, H. Yu, and B. K. Woods, "An investigation into 3D printing of fibre reinforced thermoplastic composites," *Additive Manufacturing*, vol. 22, pp. 176–186, 2018, doi: 10.1016/j.addma.2018.04.039.

[5] Y. Tao, H. Wang, Z. Li, P. Li, and S. Q. Shi, "Development and application of wood flour-filled polylactic acid composite filament for 3D printing," *Materials*, vol. 10, no. 4, p. 339, 2017, doi: 10.3390/ma10040339.

[6] C. Wang, L. M. Smith, W. Zhang, M. Li, G. Wang, S. Q. Shi, H. Cheng, and S. Zhang, "Reinforcement of polylactic acid for fused deposition modeling process with nano particles treated bamboo powder," *Polymers*, vol. 11, no. 7, p. 1146, 2019, doi: 10.3390/polym11071146.

[7] A. P. Mathew, K. Oksman, and M. Sain, "Mechanical properties of biodegradable composites from poly lactic acid (PLA) and microcrystalline cellulose (MCC)," *Journal of Applied Polymer Science*, vol. 97, no. 5, pp. 2014–2025, 2005, doi: 10.1002/app.21779.

[8] C. A. Murphy and M. N. Collins, "Microcrystalline cellulose reinforced polylactic acid biocomposite filaments for 3D printing," *Polymer Composites*, vol. 39, no. 4, pp. 1311–1320, 2018, doi: 10.1002/pc.24069.

[9] Q. Wang, C. Ji, J. Sun, Q. Yao, J. Liu, R. M. Y. Saeed, and Q. Zhu, "Kinetic thermal behavior of nanocellulose filled polylactic acid filament for fused filament fabrication 3D printing," *Journal of Applied Polymer Science*, vol. 137, no. 7, 2020, Art. no. 48374, doi:10.1002/app.48374.

[10] N. D. Ahmad and M. W. Wildan, "Preparation and properties of cellulose nanocrystals-reinforced Poly (lactic acid) composite filaments for 3D printing applications," *Results in Engineering*, vol. 17, 2023, Art. no. 100842, doi: 10.1016/j.rineng.2022.100842.

[11] M. Vinyas, S. J. Athul, D. Harursampath, and T. N. Thoi, "Experimental evaluation of the mechanical and thermal properties of 3D printed PLA and its composites," *Materials Research Express*, vol. 6, no. 11, 2019, Art. no. 115301, doi: 10.1088/2053-1591/ab43ab.

[12] P. Kakanuru and K. Pochiraju, "Moisture ingress and degradation of additively manufactured PLA, ABS and PLA/SiC composite parts", *Additive Manufacturing*, vol. 36, 2020, Art. no. 101529, doi: 10.1016/j.addma.2020.101529.

[13] T. N. M. Irfan, T. S. George, K. S. Abidh, S. Prakash, B. P. Kanoth, N. George, V. B. Kurup, C. D. M. Dominic, and A. B. Nair, "Waste paper as a viable sustainable source for cellulosic extraction by chlorine free bleaching and acid hydrolysis method for the production of PVA-starch/cellulose based biocomposites," *Materials Today: Proceedings*, 2023, doi: 10.1016/j.matpr. 2023.03.805.

[14] M. Kam, A. Ipekci, and Ö. Şengül, "Investigation of the effect of FDM process parameters on mechanical properties of 3D printed PA12 samples

using Taguchi method," *Journal of Thermoplastic Composite Materials*, vol. 36, no. 1, pp. 307–325, 2023, doi: 10.1177/08927057211006459.

[15] N. Ayrilmis, M. Kariz, J. H. Kwon, and M. K. Kuzman, "Effect of printing layer thickness on water absorption and mechanical properties of 3D-printed wood/PLA composite materials," *The International Journal of Advanced Manufacturing Technology*, vol. 102, pp. 2195–2200, 2019, doi: 10.1007/s00170-019-03299-9.

[16] W. Wu, P. Geng, G. Li, D. Zhao, H. Zhang and J. Zhao, "Influence of layer thickness and raster angle on the mechanical properties of 3D-printed PEEK and a comparative mechanical study between PEEK and ABS," *Materials*, vol. 8, no. 9, pp. 5834–5846, 2015, doi: 10.3390/ma8095271.

[17] B. Coppola, N. Cappetti, L. Di Maio, P. Scarfato, and L. Incarnato, "3D printing of PLA/clay nanocomposites: Influence of printing temperature on printed samples properties," *Materials*, vol. 11, no. 10, p. 1947, 2018, doi: 10.3390/ma11101947.

[18] P. Parandoush and D. Lin, "A review on additive manufacturing of polymer-fiber composites," *Composite Structures*, vol. 182, pp. 36–53, 2017, doi: 10.1016/j.compstruct.2017.08.088.

[19] S. F. Kabir, K. Mathur, and A. F. M. Seyam, "Maximizing the performance of 3d printed fiber-reinforced composites," *Journal of Composites Science*, vol. 5, no. 5, p. 136, 2021, doi: 10.3390/jcs5050136.

[20] T. Anto, R. C. Rajendran, P. K. Muraleedharan, and E. Jayamani, "Effect of borax-boric acid treatment on fire resistance, thermal stability, acoustic, and mechanical properties of mycelium bio composites," *Applied Science and Engineering Progress*, vol. 17, no. 4, 2024, Art. no. 7271, doi: 10.14416/j.asep.2023.11.007.

[21] S. Phongtamrug, P. Pilasen, and K. Ridthitid, "Effects of silane coupling agents on physical properties of simultaneous biaxially stretched polylactide film," *Applied Science and Engineering Progress*, vol. 17, no. 3, p. 7406, 2024, doi: 10.14416/j.asep.2024.06.012.

[22] A. Dogru, A. Sozen, G. Neser, and M. O. Seydibeyoglu, "Effects of aging and infill pattern on mechanical properties of hemp reinforced PLA composite produced by fused filament fabrication (FFF)," *Applied Science and Engineering Progress*, vol. 14, no. 4, pp. 651–660, 2021, doi: 10.14416/j.asep.2021.08.007.

[23] M. Y. Kim, C. Kim, J. Moon, J. Heo, S. P. Jung, and J. R. Kim, "Polymer film-based screening and isolation of polylactic acid (PLA)-degrading microorganisms," *Journal of Microbiology and Biotechnology*, vol. 27, no. 2, pp. 342–349, 2017, doi: 10.4014/jmb.1610.10015.

[24] A. F. Alwan, "Preparation and characterization of polylactic acid by ring opening polymerization using unconventional heating system," *Al-Mustansiriyah Journal of Science*, vol. 33, no. 2, pp. 24–30, 2022, doi: 10.23851/mjs.v33i2.1119.

[25] S. Huang, J. Hwang, H. Liu, and A. Zheng, "A characteristic study of polylactic acid/organic modified montmorillonite (PLA/OMMT) nanocomposite materials after hydrolyzing", *Crystals*, vol. 11, no. 4, p. 376, 2021, doi: 10.3390/cryst11040376.

[26] Q. Wang, C. Ji, J. Sun, Q. Zhu, and J. Liu, "Structure and properties of polylactic acid biocomposite films reinforced with cellulose nanofibrils," *Molecules*, vol. 25, no. 14, p. 3306, 2020, doi: 10.3390/molecules25143306.