

The Influence of coffee grounds waste on the thermal and mechanical characteristics of polymer-based 3D printing filament materials

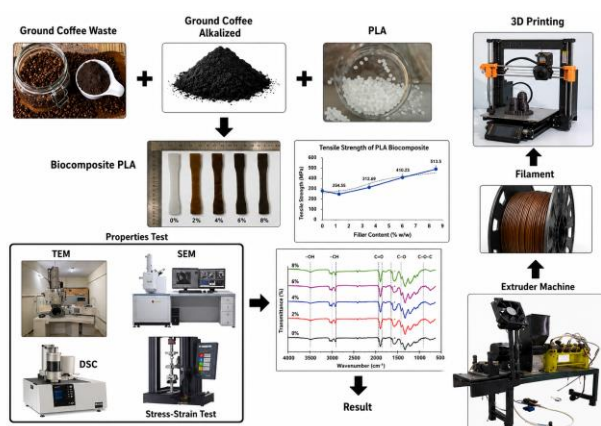
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Highlights:

- Alkali-treated coffee grounds were successfully utilized as a sustainable PLA reinforcement.
- Mechanical properties improved progressively with increasing coffee grounds filler content.
- Thermal performance of PLA biocomposites increased through enhanced filler–matrix interaction.

Abstract

The development of polymer-based 3D printing filaments is challenged by the need for greater mechanical strength and thermal stability. Coffee grounds waste was selected as a filler to enhance the properties of polylactic acid (PLA), a widely used polymer filament. The coffee grounds were treated with NaOH to remove lignin, hemicellulose, and impurities, thereby improving their compatibility with the PLA matrix. The treated filler was incorporated into the polymer matrix at 2%- 8% w/w and molded into standard tensile test specimens according to ASTM D638. Mechanical testing and thermal analysis were performed to assess the performance of the resulting biocomposites. Results indicate that increasing the filler content in 2% w/w increments up to 8% w/w enhances tensile strength by approximately 1.5 MPa, tensile strain by 0.002, and elastic modulus by 30.378 MPa. Thermal analysis also shows an increase in glass transition temperature (T_g) from 59 °C to 66 °C and a rise in degradation peak temperature from 335.9 °C to 371.1 °C. These enhancements are attributed to improved interfacial adhesion between the treated filler and the PLA matrix, which restricts polymer chain mobility. Alkali-treated coffee grounds waste thus represents a promising sustainable filler for enhancing the mechanical and thermal performance of polymer-based 3D printing filaments.

Keywords: Coffee; Alkalized; Waste; Filament; 3D printing

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1. Introduction

3D printing, also known as additive manufacturing, is a digital manufacturing process that offers almost unlimited opportunities to develop structures or objects by adjusting material composition, processing conditions, and geometry at every point in an object [1]. 3D printing technology uses filament material that is melted through a nozzle and moved according to commands from CAD software to create 3D models layer by layer. The 3D printing process, which can quickly build products with complex geometric shapes, contributes to the growth of the market sector [2]. 3D printing has also revolutionized production processes by making them more efficient,

reducing waste, and enabling on-demand, customized manufacturing [3]. The use of 3D printing technology has become increasingly common in the automotive industry for designing new components and reproducing existing ones through digitization, reverse engineering, CAD model creation, and prototype fabrication [4]. The implementation of various materials in 3D printing technology, such as polymers, metals [5], chocolate [6], cement [7], biomaterials [8], and others, has encouraged the use of this technology in fields including health, biomedical [9], pharmaceutical [10], food, and building construction, with the potential to be developed in many other areas. The ability of 3D printing to build products using a variety of materials makes it a popular choice, especially for building innovative product designs. The development of this technology in manufacturing, as well as the methods and materials used in product construction, presents numerous opportunities for further improvement.

The material used in 3D printing is in the form of a filament that is rolled onto a roller. The filament will be melted and ejected through a moving nozzle to print the product according to the commands from the CAD software and its slicer. The accuracy of the geometry of components produced by the 3D printing extrusion process depends on both the product design and the process parameters [11], and the properties of the raw material filament [12]. Therefore, the filament material in 3D printing plays a vital role in the quality of the product built [13]. PLA, ABS, and PETG filaments are widely used because they are readily available [14]. PLA is the most dominant because it is the cheapest and easiest to buy. PLA is a hydrophilic biopolymer that readily absorbs water and decomposes naturally; however, it exhibits weaknesses due to its low mechanical and thermal properties [15], [16], [17]. Several studies have been conducted to improve the properties of PLA by compounding natural fibers as fillers into the polymer matrix [18], [19].

Natural fibers from coffee are also being studied as potential fillers. They can be used in bio-polymer-based matrices, such as PLA, or synthetic polymers, including HDPE, ABS, PP, PET, and others. The use of coffee grounds as fillers has attracted attention due to their abundance. Coffee grounds are a waste product from daily coffee consumption and pose a disposal problem. Recycling is also an alternative to reduce the number of clogged drains caused by coffee waste. Several studies have found that using coffee grounds waste directly, without prior treatment, reduces their tensile strength and tensile strain, although it can increase their Young's modulus [20], [21]. The alkalinizing process for coffee grounds waste filler aims to roughen the surface of the grounds, thereby improving adhesion and enhancing bonding to the polymer matrix. This study will investigate whether alkalinized coffee grounds waste can enhance the mechanical and thermal properties of PLA. The research is a preliminary study, so the specimens were prepared in accordance with ASTM D638 using a laboratory-scale molding method. The chosen method is based on previous studies reporting that mechanical property test results for specimens produced by injection molding and 3D printing are similar [22], [23].

2. Method

The implementation of this research involves several steps, namely: preparation of tools, preparation of polymer matrix materials and filler materials, printing of specimens according to the ASTM D 638 standard form, taking mechanical test data, thermal and FTIR (Fourier Transformation Infra-Red) testing, processing of test result data, analysis of biocomposites characteristics, discussion, conclusion and preparation of reports and publications. The process flow for making PLA bio-composite filaments, from specimen preparation through testing and utilization in 3D printing, is shown in Figure 1.

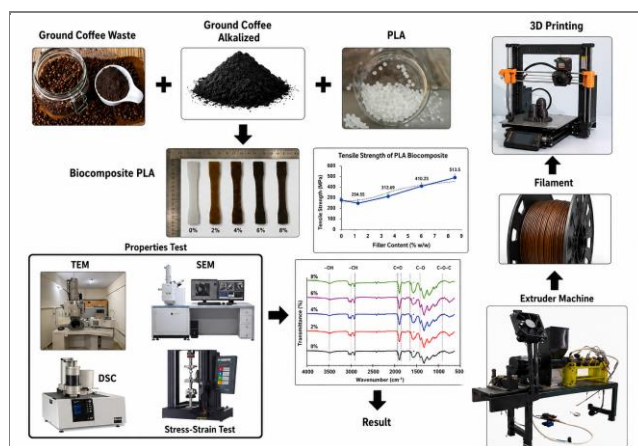


Figure 1.
The flow process of
3D printing

2.1. Machine and Mold Preparation

The tools required to create the test specimens are test specimen molds that follow the geometry specified in ASTM D638, as illustrated in Figure 2. Two molds are made of an Aluminium

alloy and machined on a CNC milling machine to maintain precision. After the molds are prepared, the oven and its equipment are prepared and calibrated.

2.2. Preparation Specimen ASTM D638

Coffee grounds waste in the bio-composites will be alkaliized first using NaOH before being used as a filler. The process is necessary because of the amorphous hemicellulose content, which leads to poor interfacial bonding with the polymer matrix. Alkali treatment using NaOH is commonly applied to overcome this issue, as it disrupts hydrogen bonds, removes lignin and other non-cellulosic components, and increases surface roughness. These changes improve cellulose exposure and interfacial adhesion, leading to better filler dispersion and enhanced bio-composite properties [24].

Figure 2.
Test specimen
according to ASTM
D638 [25]

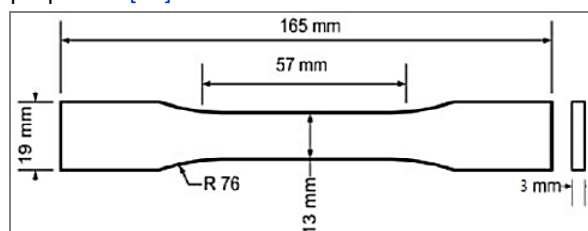


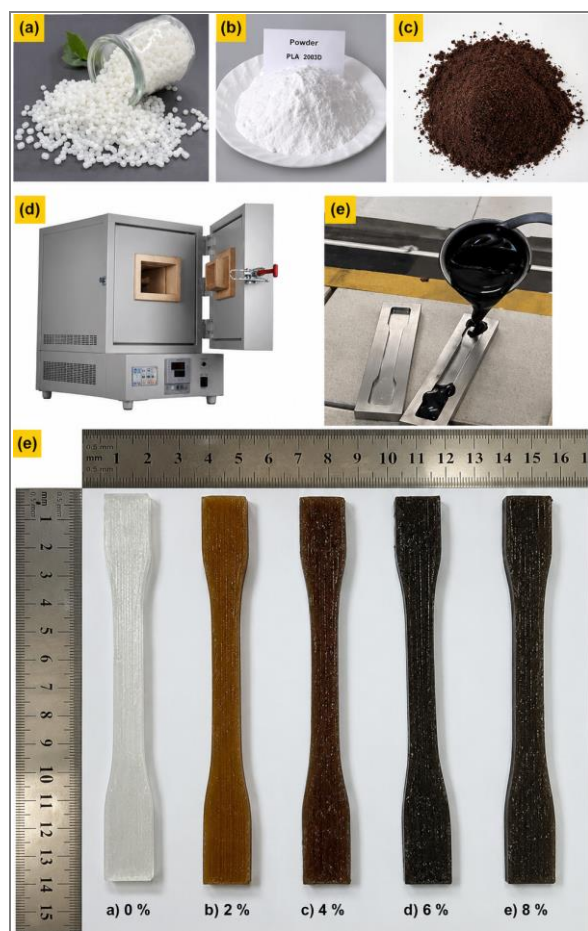
Figure 3.
Alkalisation process of
filler



The alkalization step begins with washing the ground coffee to remove impurities, followed by soaking it in a 5% NaOH solution at room temperature for 2 hours. After that, it is washed thoroughly with distilled water until clean, and the pH is approximately 7. Then, it is dried in an oven at 80 °C and cooled. The coffee grounds waste is then filtered, and only particles with a grain size above 80 mesh are used as a filler to facilitate mixing. The alkalization process of coffee grounds waste is illustrated in Figure 3.

The matrix material is a raw material derived from PLA filament, which is commonly available in the local market because it is inexpensive and readily available through online marketplaces. This consideration is intended to facilitate industrial-scale production of this biocomposite filament in the future. However, the PLA material used is not standardized, so its mechanical and thermal properties need to be tested. Previous research on the effect of layer height on the tensile strength of 3D-printed products using ASTM D 638 specimens made of PLA filament from a well-known brand in Indonesia also reported a tensile strength range of 7.07 to 8.59 MPa, which is well below the theoretical tensile strength of PLA [26]. The mechanical properties of this PLA material without filler will be tested first as a reference for comparing changes in the characteristics of bio-composites. PEG will be used as a compatibilizer to enhance interfacial interactions, as it possesses hydroxyl (-OH) groups that can interact with polar groups on the surfaces of natural fibers, thereby improving the dispersion of particles or fibers within the polymer matrix. Additionally, PEG also serves as a plasticizer, making the polymer chain more flexible and mobile. The PEG content is 10% w/w for each weight fraction of the specimen [27]. The weight of the matrix material,

Figure 4.
The specimen
preparation process:
(a) Matrix;
(b) PEG;
(c) Alkaliized Coffee
Grounds Waste;
(d) Melting;
(e) Molding;
(f) Specimen



compatibilizer, and filler is calculated by considering the shrinkage factor. The mold volume is used to convert to weight, and the weight fraction of each specimen material is calculated. PLA is melted at 200 °C and maintained at that temperature for 2 minutes, then PEG and coffee ground powder are added to the PLA melt, which is stirred at 100 rpm for 10 minutes. The mixture is then poured into a specimen mold, which was subsequently closed and allowed to cool at room temperature. After 30 minutes, the mold is opened, and the PLA biocomposite test specimen is removed. The process is illustrated in Figure 4.

2.3. Methods of Bio-composites Properties Test

After the specimen is ready, its mechanical properties are tested, including tensile strength, tensile strain, and elastic modulus. FTIR testing and SEM are used to determine the nature of chemical bonds and functional groups of each component. TGA testing is also conducted to determine its thermal properties, including glass transition temperature (T_g), temperature stability, and the temperature at which degradation begins.

3. Results and Discussion

This research focuses on the potential to produce 3D printing filaments from PLA bio-composites using alkalized ground coffee as a filler to enhance the properties of existing PLA filaments. Tests were conducted on ASTM D638 standard specimens to determine the mechanical and thermal properties of the bio-composite material intended for filament use as a raw material for 3D printing products.

3.1. Fourier Transform Infra-Red Spectroscopy (FTIR)

The FTIR spectrum of filler (coffee grounds waste) and PLA composites in Figure 5 clearly demonstrates the incorporation of coffee particles into the polymer matrix through several characteristic absorption features. The FTIR spectrum in Figure 5 confirms the incorporation of coffee grounds waste filler into the PLA matrix through several characteristic absorption bands. A broad O-H stretching peak at $\sim 3421\text{ cm}^{-1}$ (0% w/w) shifts to $\sim 3388\text{ cm}^{-1}$ for 2-8% w/w and increases in intensity, indicating hydroxyl groups from lignocellulosic coffee components and absorbed moisture. The aliphatic C-H stretching at ~ 2919 to $\sim 2921\text{ cm}^{-1}$ shows slight changes, suggesting interaction between polymer chains and filler. The strongest C=O peak at ~ 1746 to $\sim 1750\text{ cm}^{-1}$ remains dominant but exhibits minor shifts, indicating possible hydrogen bonding between PLA carbonyl groups and coffee hydroxyl groups. CH₂ bending at ~ 1452 to $\sim 1442\text{ cm}^{-1}$ and C-O-C stretching at ~ 1055 to $\sim 1081\text{ cm}^{-1}$ show small intensity and position variations, reflecting modified interfacial bonding and structural rearrangement after filler addition. Peaks near 755 cm^{-1} become more pronounced with increasing filler content, confirming the presence of aromatic lignin structures from coffee. These changes verify effective compounding of coffee filler in PLA, although the increased O-H absorption suggests reduced hydrophobicity of the bio-composite.

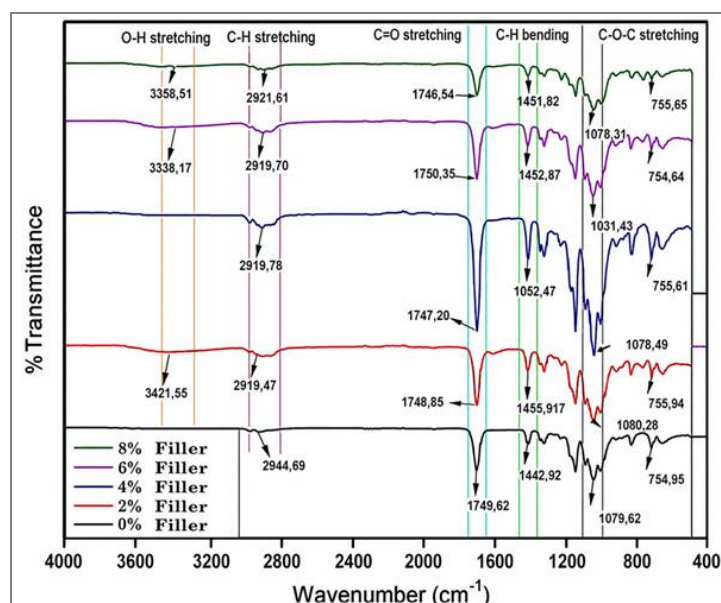
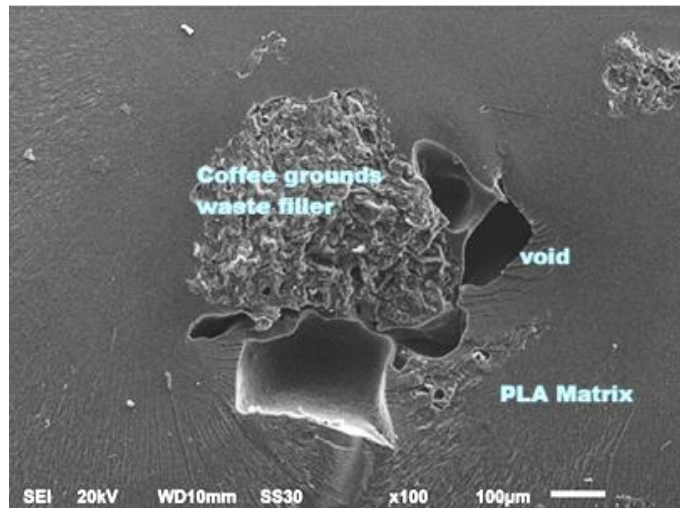


Figure 5.
FTIR test result for the
filaments of bio-
composite

3.2. Scanning Electron Microscope (SEM) Test

Morphological analysis using a Scanning Electron Microscope (SEM) was conducted on the fracture surface of the PLA bio-composite specimen containing 8% w/w coffee grounds waste filler, which exhibited the highest tensile strength among all tested compositions.

Figure 6.
SEM image at 100×
magnification on the
fracture cross-section of
the specimen with 8%
filler concentration



The SEM image at 100× magnification in **Figure 6** demonstrates a non-homogeneous fracture surface, with filler particles dispersed throughout the PLA matrix. Despite this dispersion, the agglomeration of irregularly shaped, rough-surfaced particles remains evident. Several voids and gaps are present around the filler, suggesting debonding resulting from imperfect interfacial adhesion between the matrix and the filler. In certain regions,

holes left by detached particles are visible, indicating a pull-out mechanism where filler particles were released from the matrix during tensile loading. The rough surface of the filler facilitates mechanical interlocking, enabling the 8% w/w composition to achieve the highest tensile strength despite micro-voids and agglomeration. The disparity in properties between the hydrophobic PLA matrix and the more hydrophilic lignocellulosic filler derived from coffee grounds waste likely contributes to the formation of micro-voids during tensile loading. These voids and micro-voids serve as stress concentration points and can initiate crack propagation under tensile stress. The relatively smooth fracture surface of the matrix and the presence of sharp cracks are indicative of the brittle fracture behavior characteristic of PLA, where failure occurs after reaching maximum stress with minimal plastic deformation.

3.3. Mechanical Properties Test

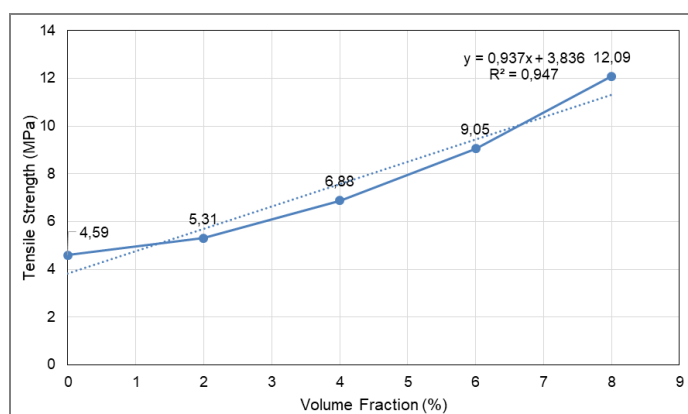
The addition of weight-fraction coffee-ground waste bio-composites, as shown in **Figure 7**, illustrates the relationship between coffee waste filler weight fraction and the bio-composites' tensile strength (MPa). Each tensile strength test for the weight fraction of coffee grounds waste filler specimens was conducted in five repetitions to ensure validity, and the results were statistically analyzed to obtain the standard deviation. For specimens with 0% w/w filler, the tensile strength was 4.59 ± 0.95 MPa; at 2% w/w, it increased to 5.31 ± 0.56 MPa; at 4% w/w, it reached 6.88 ± 0.93 MPa; at 6% w/w, it further increased to 9.05 ± 0.99 MPa; and at 8% w/w, the tensile strength achieved 12.09 ± 1.69 MPa.

This indicates that incorporating filler significantly enhances the mechanical properties of the PLA matrix. The linear regression equation ($y = 0.937x + 3.836$) with a high coefficient of determination ($R^2 = 0.947$) suggests a strong linear correlation between filler content and tensile strength. Overall, the results demonstrate that the reinforcement effect of the filler is highly effective, as the tensile strength of the bio-composite nearly tripled when the filler content increased from 0% to 8%. This confirms the positive effect of filler incorporation on the load-bearing capacity of the PLA-based composite.

The next mechanical property of a bio-composite to be discussed is tensile strain, a measure of the relative change in length of a material under an applied tensile force. Tensile strain is the

change in length of the bio-composite specimen with varying weight fractions from 0% w/w to 8% w/w, divided by its original length, indicating how much the material has stretched or elongated. **Figure 8** illustrates the tensile strain test results for the bio-composites' mechanical properties. The initial condition of pure PLA without filler (0%) exhibits a tensile strain of 0.016 mm/mm,

Figure 7.
The effect of weight
fraction filler on PLA
bio-composites tensile
strength



which will be used as the baseline ductility in observing the effect of filler addition on the bio-composites.

The addition of filler at low weight fractions (2% w/w–4% w/w) increases the strain to 0.021–0.022 mm/mm, indicating improved ductility due to a more uniform stress distribution and stronger interfacial bonding. The strain at an intermediate loading (6% w/w) increases slightly to 0.023 mm/mm. This condition indicates a transition in which the reinforcing effect begins to limit the mobility of the polymer chains, thereby balancing stiffness and toughness. In the bio-composites with a higher filler content (8%w/w), a maximum strain of 0.026 mm/mm is achieved, indicating that the composite retains its ductility due to good filler dispersion and strong interfacial adhesion, which helps prevent premature crack initiation.

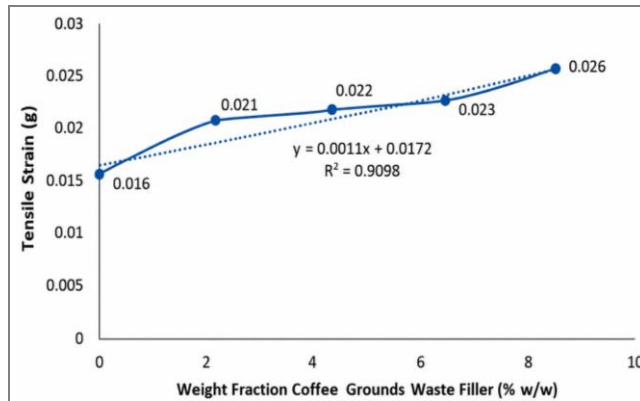


Figure 8.
The effect of weight fraction filler on PLA bio-composites tensile strain

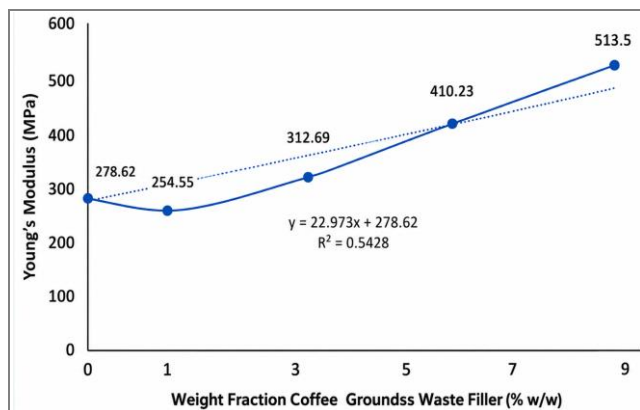


Figure 9.
The effect of weight fraction filler on PLA bio-composites Young's modulus

The Young's Modulus of the PLA bio-composite showed a nonlinear dependence on filler weight fraction, indicating a complex interaction between the filler and the polymer matrix. PLA with 0% filler had a modulus of 278.62 MPa, but decreased sharply to 254.55 MPa at 2% and 312.69 MPa at 4%, indicating that low filler loading may disrupt the continuity of the PLA matrix or cause inadequate stress transfer due to weak interfacial adhesion or filler agglomeration. However, above 4% loading, the modulus increased substantially, reaching 410.23 MPa at 6% and peaking at 513.50 MPa at 8%, reflecting better filler dispersion and stronger interfacial bonding at higher concentrations. The equation of the curve, $y = 22.973x + 278.62$, with $R^2 = 0.5428$, shows only a moderate correlation and is not entirely linear; it is influenced by competing filler reinforcement and matrix disruption mechanisms, as illustrated in Figure 9.

3.4. Thermal Properties Test

Thermogravimetric analysis (TGA) curves in Figure 10 show that PLA biocomposites become more thermally stable as coffee waste filler content increases, as indicated by higher onset and endset

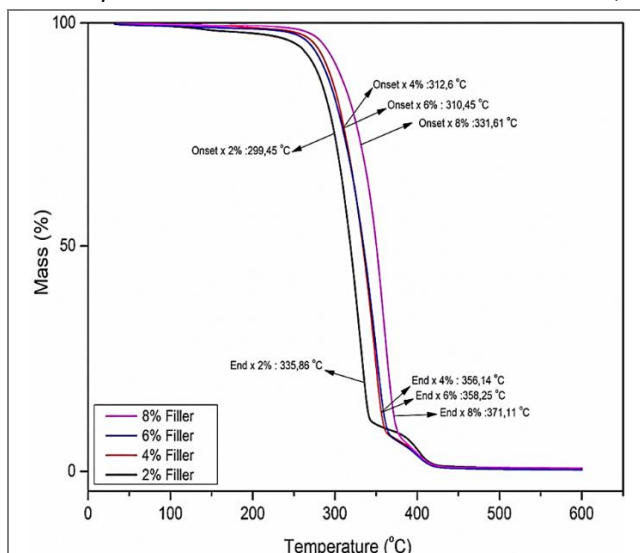


Figure 10.
The TGA test result for specimens with 2%, 4%, 6%, and 8% filler concentration

decomposition temperatures. At 2% w/w filler, the onset and endset temperatures are 299.45 °C and 335.86 °C, representing the lowest thermal resistance. Increasing the filler content to 4% and 6% w/w yields onset temperatures of 312.6 °C and 310.45 °C, and endset temperatures of 356.14 °C and 358.25 °C, indicating improved thermal stability relative to the 2% w/w composite. The highest thermal resistance occurs at 8% w/w filler, with an onset temperature of 331.61 °C and a much higher endset temperature of 371.11 °C, indicating a significant delay in thermal degradation.

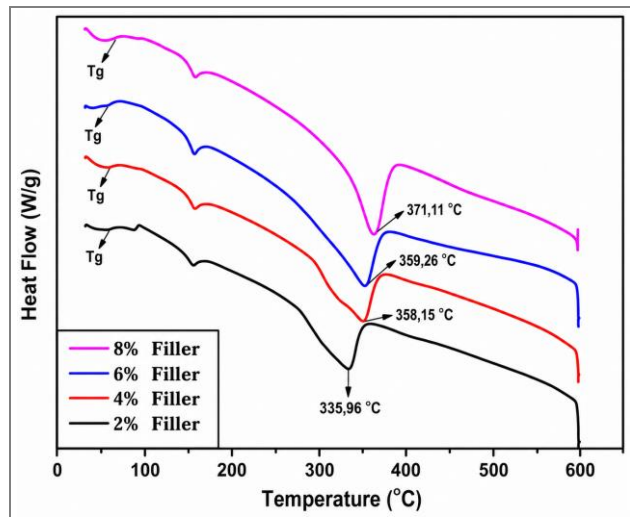


Figure 11.
The DSC test result for
specimens with 2%,
4%, 6%, and 8% filler
concentration

The increases in onset and endset temperatures indicate that the coffee grounds waste filler improves the composite's thermal resistance by restricting polymer chain mobility, strengthening interfacial interactions between the PLA matrix and the filler, and promoting the formation of a stable coffee-char layer during decomposition. This char layer on the coffee grounds waste acts as a barrier to heat transfer and volatile diffusion, reducing the degradation rate and further improving the thermal stability of the PLA bio-composite.

Differential Scanning Calorimetry (DSC) thermograms in [Figure 11](#) show the effect of filler addition on the thermal transition and stability of PLA bio-composite. All specimens exhibited a glass transition temperature (T_g), followed by a central degradation peak at higher temperatures. At a low filler content (2% w/w), the degradation peak occurred at 335.86 °C, indicating the lowest thermal resistance. When the filler weight fraction increased to 4% w/w and 6% w/w, the degradation temperatures shifted to 356.15 °C and 358.26 °C, respectively, reflecting improved thermal stability due to better interfacial interactions that hinder polymer chain mobility. The highest stability was achieved at an 8% w/w filler concentration, with a peak temperature of 371.11 °C, indicating strong filler-matrix bonding and a practical insulating effect that delayed thermal decomposition. Overall, the results suggest that higher filler additions improve the thermal resistance of the PLA matrix by increasing chain stiffness and promoting thermal insulation.

3.5. Discussion

The primary limitation of polylactic acid (PLA) as a 3D printing filament is its relatively low mechanical strength and thermal resistance. Previous studies have shown that incorporating untreated coffee grounds waste into PLA reduces tensile strength, primarily due to weak interfacial bonding between the hydrophobic polymer matrix and the hydrophilic lignocellulosic filler [28]. In the present study, alkaline treatment was employed to enhance the filler's surface activity and its compatibility with PLA. Fourier-transform infrared spectroscopy (FTIR) results indicate an increase in O–H absorption intensity and slight shifts in the C=O peak, which suggest stronger hydrogen bonding between lignocellulosic hydroxyl groups and PLA carbonyl groups following alkalization. Scanning electron microscopy (SEM) observations corroborate these findings by revealing improved filler embedding and mechanical interlocking. However, some voids and pull-out features persist, indicating that while adhesion is enhanced, it is not completely uniform. Mechanical testing reveals a consistent increase in tensile strength up to 8 % w/w filler, with a strong linear correlation. This confirms that alkalization and the addition of polyethylene glycol (PEG) as a compatibilizer improve stress transfer between the matrix and the filler. The observed increase in tensile strain indicates improved stress distribution and delayed crack initiation. Modulus behavior reflects competing mechanisms: at low filler content, matrix continuity is disrupted, whereas at higher filler loading, reinforcement is more effective due to stronger interfacial bonding and improved dispersion.

Thermal analysis reveals trends consistent with the mechanical findings. Thermogravimetric analysis demonstrates that increasing filler content shifts both the onset and endset degradation temperatures to higher values, indicating enhanced thermal stability. This improvement is attributed to stronger filler-matrix interactions and the formation of a lignocellulosic char layer during decomposition, which acts as a thermal barrier by slowing heat transfer and volatile diffusion. Differential scanning calorimetry (DSC) results further confirm reduced polymer chain mobility, as evidenced by an increase in glass transition temperature. The higher degradation peak temperature suggests improved structural stability of the composite. Additionally, the filler may function as a nucleating agent, promoting the development of more stable crystalline regions within the PLA matrix. The concurrent improvements in tensile strength, glass transition temperature, and thermal resistance indicate that the alkalized coffee filler serves as an active reinforcement rather than a passive additive. Within the tested range, the 8% w/w composition

achieves an optimal balance among dispersion, interfacial adhesion, and matrix integrity, highlighting its strong potential for developing sustainable PLA bio-composite filaments for 3D printing applications.

4. Conclusion

This study demonstrates that alkali-modified coffee ground waste can be effectively utilized as a functional reinforcement in PLA-based composites for filament applications. The surface modification enhances compatibility with the polymer matrix, enabling more efficient load transfer and improving structural performance. Among the compositions evaluated, the 8% w/w filler content provides the most favorable balance between mechanical integrity and thermal durability. The combined effects of enhanced interfacial interactions, controlled polymer mobility, and a stabilized composite structure highlight the potential of this material system as a sustainable approach to improving PLA performance in additive manufacturing.

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Authors' Declaration

Authors' contributions and responsibilities – The authors made substantial contributions to the study's conception and design. The authors took responsibility for data analysis, interpretation, and discussion of results. The authors read and approved the final manuscript.

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Availability of data and materials – All data is available from the authors.

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