



Investigation on mechanical and vibration behavior of FDM 3D printed PLA/biochar filler biocomposites

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Abstract

The capacity to quickly create customised biopolymer-based composite components has made fused deposition modelling (FDM) a subject of great interest. In this study, poly lactic acid (PLA) and composites made from almond shell biochar were combined to create biocomposite filaments, which were then 3D printed using FDM. To enhance the performance of PLA, different weight percentages of sub-micrometric almond shell biochar particles (0%, 1%, 3%, 5%, and 10%) were added to the filament preparation process. PLA filled with 5% almond shell biochar particles demonstrated higher tensile, and flexural measuring 61 MPa and 64 MPa, respectively, compared to clean PLA, which had the lowest values. The interaction of the biochar filler, which has a strong load-carrying capacity, with the PLA matrix improved the mechanical strength of the biocomposite. The PLA biocomposite containing 5% almond shell biochar particles had a natural frequency and damping ratio of 144 Hz and 0.67, respectively. The damping of PLA filled with 5% almond shell biochar particles was found to be higher than that of neat PLA due to the inherent porous structure of the natural biochar filler. This study suggests that almond shells derived from agricultural biomass residues could serve as a sustainable material for use in PLA biocomposites, which could enhance their mechanical and vibration qualities and make them more competitive in the industry.

Keywords: Biochar particle, Poly(lactic) acid, 3D printing, Biocomposites, Mechanical properties, vibration behavior

Introduction

Poly(lactic acid) (PLA) based composites are one of the renewable thermoplastic polymers that are favored over non-biodegradable ones because of their biodegradability. These

composites have various applications such as in biomedical, automobile, electrical, agriculture and other industries [1-3]. Polymer composites derived from PLA have excellent properties including mechanical strength, flexibility, endurance, and high durability [4, 5]. Over the past few decades, additive manufacturing, also known as 3D printing, has gained popularity in the design and fabrication industries due to its low operating costs and versatility for rapid prototyping. Through the direct digital manufacturing process, a wide range of 3D products with complex geometries can be successfully built [6-8]. Additive manufacturing offers several key benefits, including the ability to produce customised and complex prototype models without the need for tooling, reduced production time, less material waste, and lower costs [9]. The high demand for functional items has prompted researchers to develop novel materials suitable for fused deposition modelling (FDM). Due to its biodegradability and environmental friendliness, polylactic acid (PLA) is the most often used raw material for FDM-based 3D printing technology. However, using pure PLA polymer in the FDM process is limited due to drawbacks such as mechanical weakness, low thermal conductivity, and water solubility rate. PLA composites, made using the appropriate AM process, have been suggested as a means of improving the quality of PLA parts produced through the FDM process [10-11].

Several studies have focused on the use of the FDM technique and natural filler reinforced PLA polymer composites for 3D printing. Various methods have been proposed to prepare PLA composite filaments, which are used as raw materials in 3D printing. However, there has been limited research on the use of biofiller and biochar PLA composites in different industries to improve the properties of 3D-printed products [12-16]. The use of biomaterials in harsh environments is a promising option. In this regard, a method to develop a biochar composite material is proposed [17]. Hydrothermal liquefaction occurs in a water medium under moderate pressure at temperatures up to 350 degrees Celsius, leading to the depolymerization of biomass and resulting in highly functionalized biochar known as hydrochar [18]. Pyrolytic processes, on the other hand, occur at temperatures above 400 degrees Celsius in limited oxygen condition. This strategy has the potential to produce biochar, non-condensable gases, and bio-oils all at once, as well as a quick and sophisticated cracking process for each biomass component, such as cellulose, lignin, and hemicellulose. [19].

Numerous studies have been conducted on the physical and mechanical properties of natural biochar composites, resulting in improved characteristics [20-23]. Poulouse et al. [24] investigated the impact of adding date palm biochar filler to polymer composites on their

mechanical, dynamic mechanical, electrical, and thermal properties. They discovered that the biochar filler boosted tensile strength and electrical conductivity. Qian et al. [25] examined the features of PLA-based composites that were mixed with bamboo biochar. Compared to pure PLA, the composites that contained biochar had better tensile, ductile, and flexural properties. The researchers found that the highest tensile strength and tensile modulus were obtained when the biochar was added up to 30%. Zhang et al. [26] studied the mechanical properties of polymer composites that were filled with bamboo biochar. The interfacial bonding between the matrix and filler was improved, resulting in better dispersion properties. The tensile and impact strength were found to be significantly enhanced due to the fine biochar filler. By increasing the percentage of particles, the mechanical characteristics of the sample were enhanced to a certain degree, but this improvement declined as the particle content in the samples increased [27, 28].

The aim of characterizing vibration and damping is to measure the impact of changes in the internal structure of adhesion between filler and matrix materials. The damping behavior has the most significant impact on filler particle variation due to the interaction in the matrix and filler reinforcement. Several research studies have shown that filler particles such as almond shell filler, fly ash, and carbon nanotubes improve the vibration properties by altering the polymer chain's structure [29-32]. Research by Kirubakaran et al. [33] examined how natural particles affected mechanical and vibrational qualities. Due to the filler reinforcement and porous structure, which increased the characteristics, it was noted that the addition of natural fillers to the polymer composite improved both mechanical and vibration behaviour.

Based on the above-mentioned discussions, there has been no previous research on the use of almond shell biochar filler filled with PLA biocomposites fabricated through the FDM process. This study aims to assess the feasibility of using FDM to create a PLA biocomposite with almond shell biochar as a filler. The study examines the mechanical properties, such as tensile and flexural as well as the vibration properties of the resulting biocomposite, to evaluate its suitability for use in bio components.

2. Materials and Method

2.1 PLA and biochar preparation

From a supplier in Chennai, India, we bought PLA (4032D) pellets. The PLA was described as having a density of 1.25 g/cc and a molecular weight of 1.8×10^5 – 2.0×10^5 g/mol in the supplier's datasheet. Biowaste almond shells were introduced into the upper chamber of a two-

step parallel auger pyrolysis reactor and heated to 500°C to produce biochar. At this temperature, the biomass underwent its initial thermo-chemical conversion, resulting in the development of biochar, which was subsequently moved to the bottom reactor, which was maintained at 800°C. N₂ flow was used to maintain an oxygen-free environment, and the conversion cycle lasted for one hour. The almond shell biochar was subsequently cleaned using a reliable method.

2.2 Fabrication process of biochar biocomposite

The PLA pellets and almond shell biochar were separately dried for 3 hours at 80°C in a curing oven. Afterward, they were blended using an industrial blender (Irweka, India) with a double cone attachment, running at 800 watts for one hour. The selection of an industrial blender is based on the size, type, and quantity of the filler being utilised. As illustrated in Table 1 shows the Compositions Wt% of biochar and PLA for biocomposites. Biochar-filled PLA composite filaments were produced using a single screw extruder with a 170°C head temperature. To create the wire filament, the screw and feed rates were set at roughly 20 and 25 rpm, respectively. Following extrusion, the PLA biocomposites were cooled, ground up, and dried. Finally, a single-screw extruder operating at 210°C generated filament with a 1.75 mm diameter.

Table 1 Compositions Wt% of biochar and PLA for biocomposites

PLA/biochar biocomposites sample designation	Biochar (Wt%)	PLA (Wt %)
PLABC0	0	100
PLABC5	1	99
PLABC10	3	97
PLABC15	5	95
PLABC20	10	90

The FDM printing technology was utilized in this study to create PLA/biochar biocomposites, and the typical FDM printing setup is depicted in Figure 1. The 3D printer specifications were as follows: Model - STRATASYSF270 with a build envelope of 305 X 254 X 305 mm, Print technology – FDM, material options - PLA, ABS, ALA, throughput

comparison - 1.5x (standard mode) and 3x (fast drafting mode), Part accuracy - 0.2 mm, and software - GrabCAD print. In addition, the following settings were made: 0°, 0 mm, 1.75 mm, ambient temperature, 0 mm, and 100% for the filling rate and the component orientation. The following conditions were used for the process: a nozzle diameter of 0.6 mm, a liquefier temperature of 220 °C, an average layer thickness of 0.1 mm, and a filling velocity of 30 mm/s.

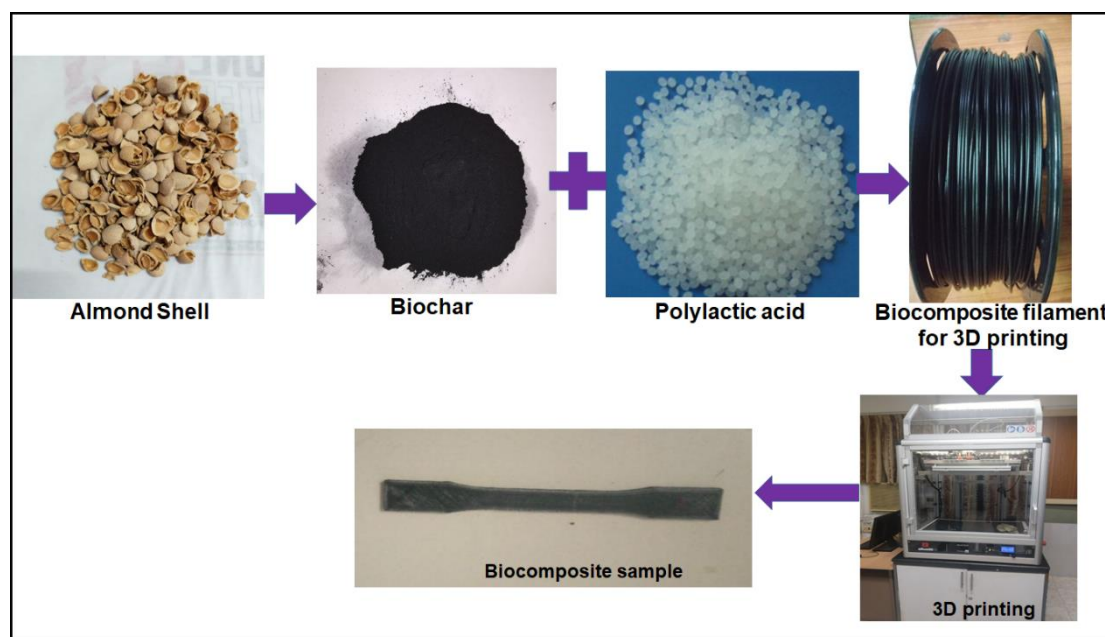


Figure 1 Fabrication process of PLA/biochar biocomposites

2.3 Tensile and flexural testing of Biochar/PLA Biocomposite

A computerised universal tensile testing machine (Make: Instron) with a crosshead speed of 1 mm/min was used to conduct tensile tests on both PLA and PLA/almond shell biochar biocomposites. The ASTM D638 standard was followed in the preparation of the tensile test samples, which had dimensions of 165*10*4 mm was printed [34]. The same universal testing apparatus was used for the flexural test, along with an additional three-point bending arrangement. Each material under test conducted a flexural test with a crosshead speed of 1 mm/min. A flexural sample with measurements of 127*12.74 mm was created in accordance with the ASTM D790 (2007) standard [34]. For each composition of the finished composite, six samples were evaluated. The standard and PLA/almond shell biochar biocomposite specimens are shown under tensile and flexural loading in Figure 2.

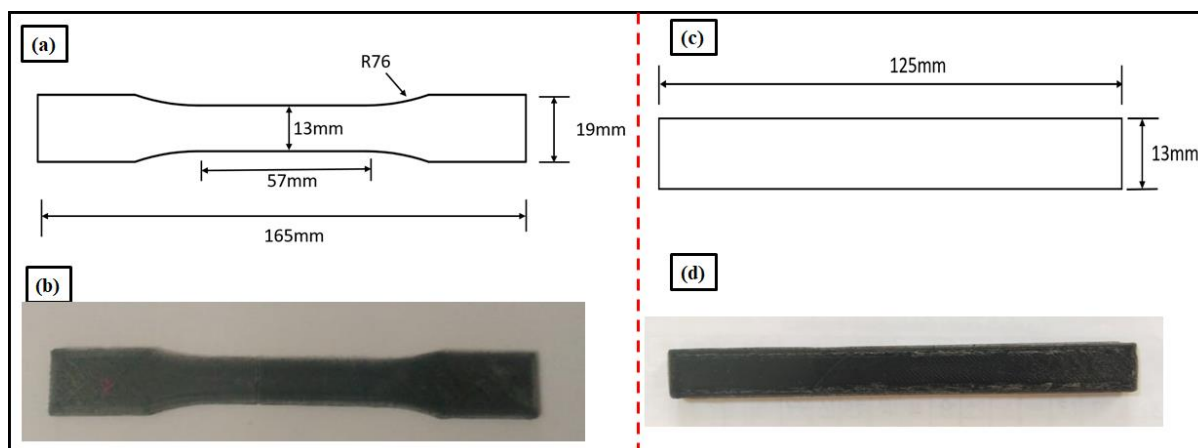


Figure 2 Standard and PLA/almond shell biochar biocomposite specimens for (a, b) Tensile loading (c, d) Flexural loading

2.4 Free vibration analysis

Using a computerised vibration shaker in cantilever beam mode, free vibration behaviour was investigated to assess the damping ratio and natural frequency of flax fibre composites filled with almond shell. The sample for the vibration test that complied with ASTM E-756 was 150 x 40 mm². The clamped laminate was excited using an impact hammer, and the vibration response of the specimen was measured using accelerometers mounted at two places on the specimen. The centre point is Point 2, which is 60 mm from the clamp end, while the free end is Point 1. The accelerometer was set to reverse while the impact hammer was slammed, and its readings were kept. In this investigation, the damping parameters were determined using the 3-dB bandwidth ($f=f_L-f_R$) method [35]. The configuration for testing the composite laminate's free vibration is shown in Figure 3. Equation 1 describes the connection between the loss factor (η), damping factor (Q), mode number (n), and damping ratio (ζ), and it is shown in Figure 3 as an embedded image. For each composition of the produced composite, six samples were examined.

$$\eta_n = \frac{1}{Q_n} = 2\zeta_n = \frac{\Delta f_{3dB}}{f_n} \quad (1)$$

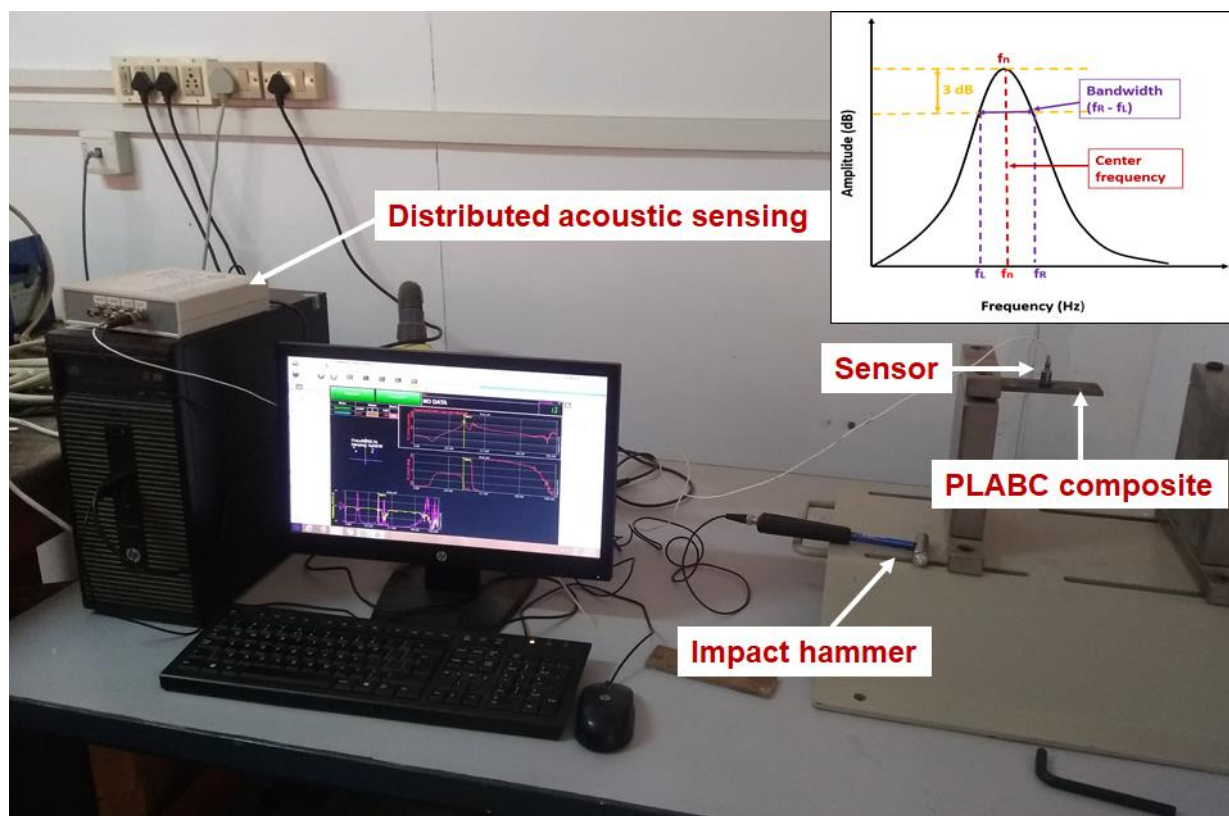


Figure 3 Vibration setup of PLA/biochar Biocomposites

3. Results and Discussion

3.1 Tensile strength of PLA/biochar Biocomposites

Figure 4 illustrates the tensile strength, stress-strain and elongation of the biocomposites composed of virgin PLA and almond shell biochar. The biocomposite's tensile strength is shown in Figure 4. The tensile strength of the biocomposite continually rises up to 5% as the concentration of almond shell biochar particle concentration rises. The PLABC5 composite obtained a peak value of 61 MPa. Tensile strength of the PLABC5 composite was 60.5% greater than that of virgin PLA. The addition of scattered almond shell biochar filler particles in the PLA matrix helped to increase the tensile strength of PLA/almond shell biochar biocomposites by sharing the tensile stress [36]. To put it another way, the extra filler particles made of almond shell biochar bonded to the PLA molecular chain, enhancing the tensile strength of the PLA/almond shell biochar biocomposite [27]. The PLA/almond shell biochar biocomposite's tensile strength was greater than that of other findings [25, 28]. The biocomposite's tensile strength, however, reduced even after adding 5% more biochar filler particles because of the poor filament quality, such as discontinuity that developed during preparation. This led to poor filling during 3D printing.

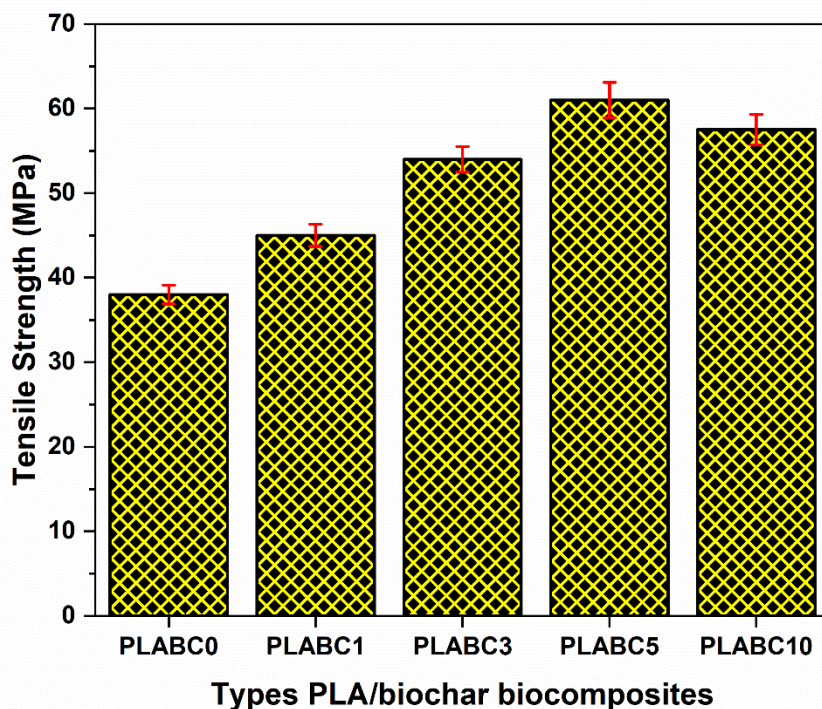


Figure 4 Tensile strength to break of PLA/biochar Biocomposites

3.2 Flexural strength of PLA/biochar Biocomposites

Figure 5 illustrates the flexural strength of PLA and its biocomposites. The flexural strength of the biocomposites continuously increased as almond shell biochar filler was added to the PLA matrix. The addition of biochar and almond shell fillers to the PLA polymer improved the flexural strength of the biocomposites. The PLABC5 and PLABC10 composites demonstrated enhanced flexural strength of 64 MPa and 60 MPa, respectively, compared to the pure PLA's 46 MPa. Comparing PLABC0 and PLABC5 composites, PLABC5 exhibited a 39.1% improvement in flexural strength. The PLA polymer and almond shell filler contributed to the composite's improvement in flexural strength due to their good compatibility and greater capacity to withstand bending stress [37, 38]. Previous research on PLA/fly and PP/landfill pine wood biocomposites also showed improved flexural strength with the addition of biochar [25, 30]. However, PLABC10 did not exhibit any improvement over the PLABC5 sample due to flaws generated during extrusion and carried over during 3D printing.

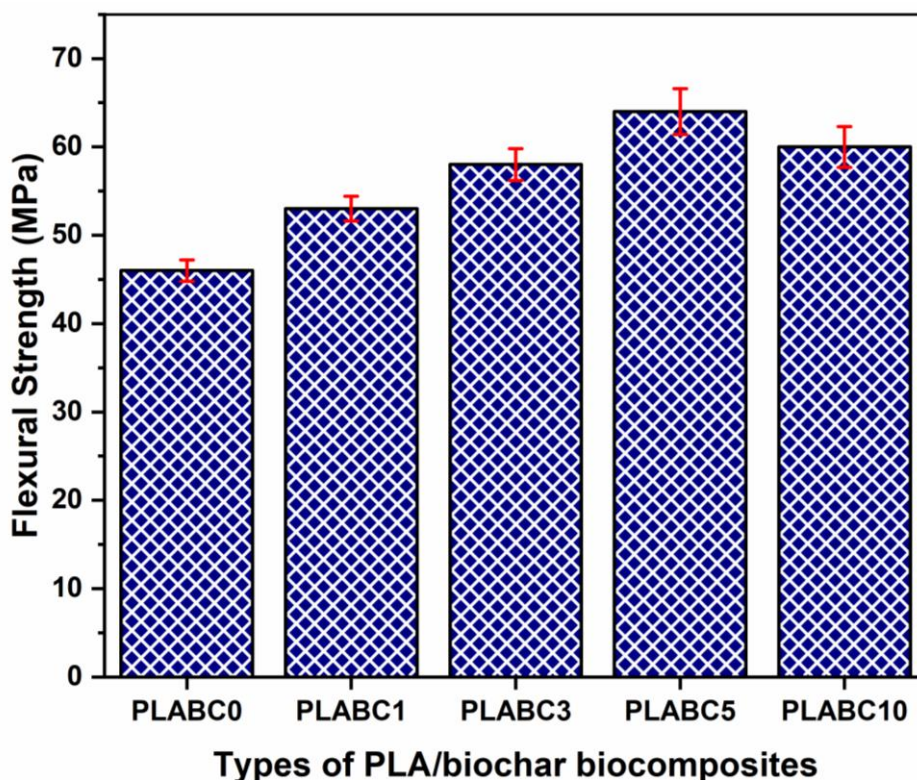


Figure 5 Flexural strength of PLA/biochar Biocomposites

3.3 Vibration and damping analysis of PLA/biochar Biocomposites

Figures 6 depict the damped natural frequencies for the first and second vibration modes, along with a summary of the information in Table 2. It can be observed from Figure 6 that adding biochar up to 5 weight percent causes the damped natural frequencies of biochar composites to increase and decrease with increasing biochar content. The maximum improvements were observed at 5 wt%, reaching 49.01% in PLA with biochar-filled composites when compared to neat PLA composites. The improvement in fundamental natural frequency was also found to be more significant than that for higher natural frequencies. As the amount of almond shell biochar in the PLA matrix increased, the second damped natural frequencies of PLA/almond shell biochar biocomposites improved up to 5% filler and then decreased [31, 33]. For example, the addition of 1%, 3%, 5%, and 10% of almond shell biochar increased the fundamental and second damped natural frequencies by 19.3%, 40.9%, 50%, and 45.4%, respectively, when compared to a neat PLA composite. A typical photocopy of natural frequency responses of mode 1 vibration for the PLABC5 of PLA/biochar biocomposites is shown in Figure 7.

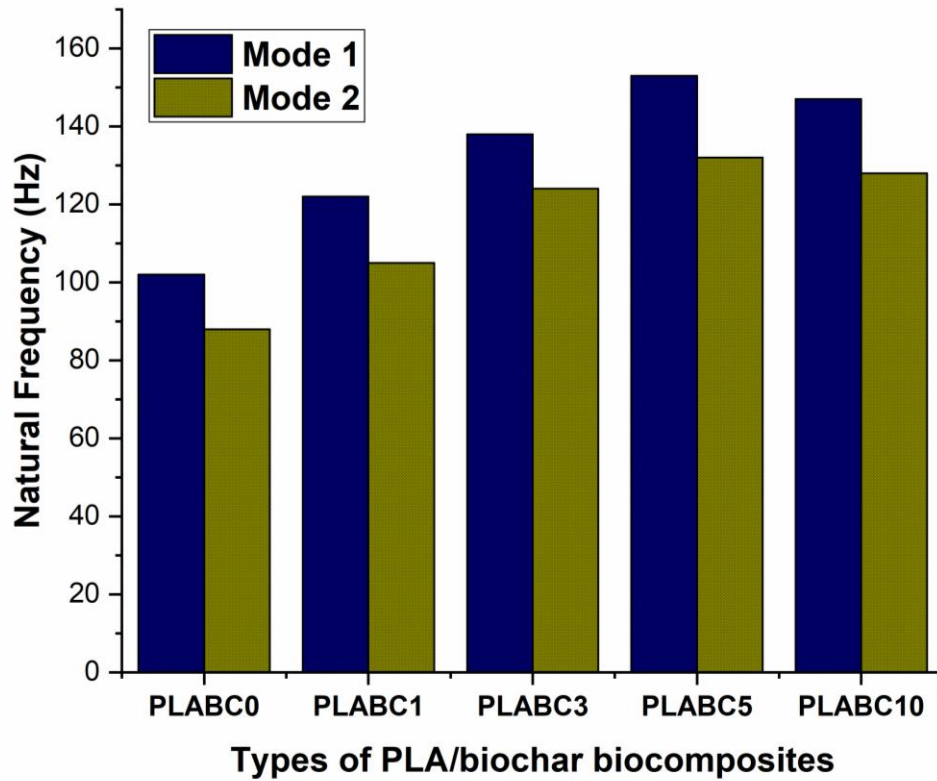


Figure 6 Natural frequency responses of mode 1 vibration for the of PLABC5 of PLA/biochar Biocomposites.

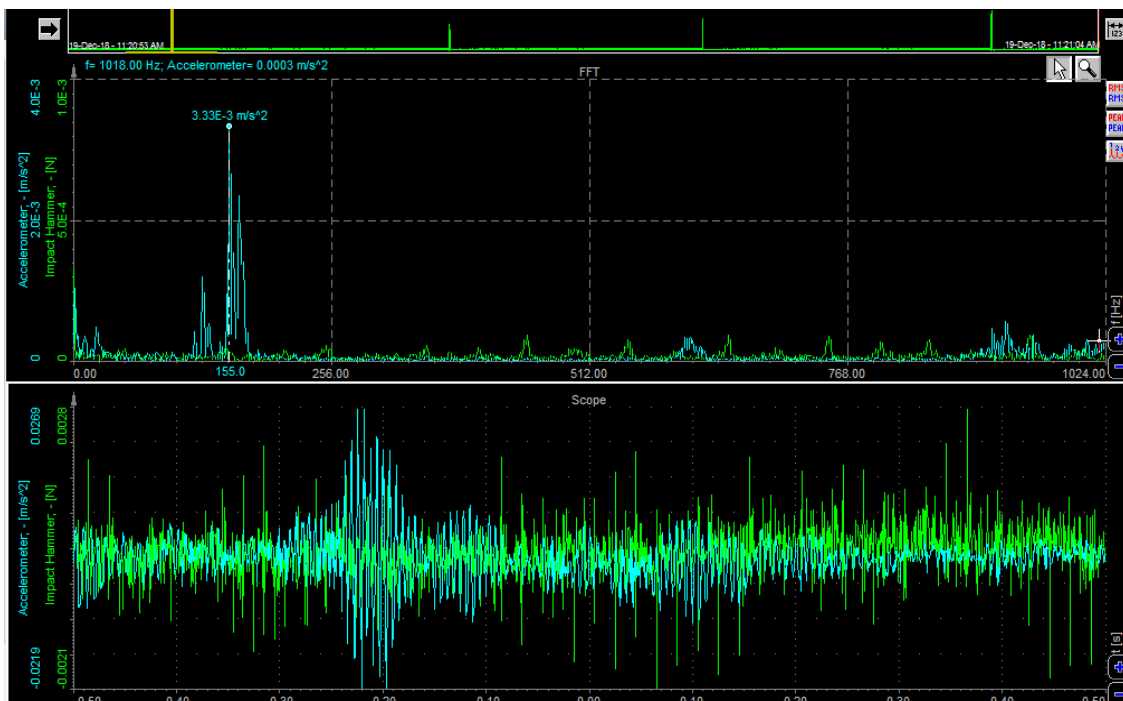


Figure 7 Typical photocopy of natural frequency response for the PLABC5 of PLA/biochar Biocomposites (mode 1)

Figure 8 illustrates the impact of biochar filler on the damping ratios of the PLA/almond shell biochar composites in mode 1 and mode 2. As shown in the figure, the addition of biochar filler enhances the damping ratios of the composites, particularly in the initial vibration modes. For instance, at a biochar filler loading of 10%, the damping ratio of the PLA/almond shell biochar biocomposites composite showed a 102% improvement. The damping ratio of biochar composites exhibits a somewhat monotonic increase. Moreover, the damping ratios of a biochar-filled biocomposite at a 10-weight percent loading showed the greatest improvement for the first mode of vibration when compared to all other biocomposites. The interaction between the filler and polymer resulted in improved vibration properties due to the natural filler reinforcement [33]. Similar improvements were observed in the damping properties of the biochar-filled biocomposite in the second mode of vibration. For example, a biochar-filled biocomposite with a 1% loading showed an improvement of over 78.5% in the damping ratio. At a loading of 10 weight percent, the biochar-filled biocomposite composites demonstrated the highest improvement in damping ratio by 140.8%. Thus, there was a remarkable improvement in the damping ratio of the second mode of vibration.

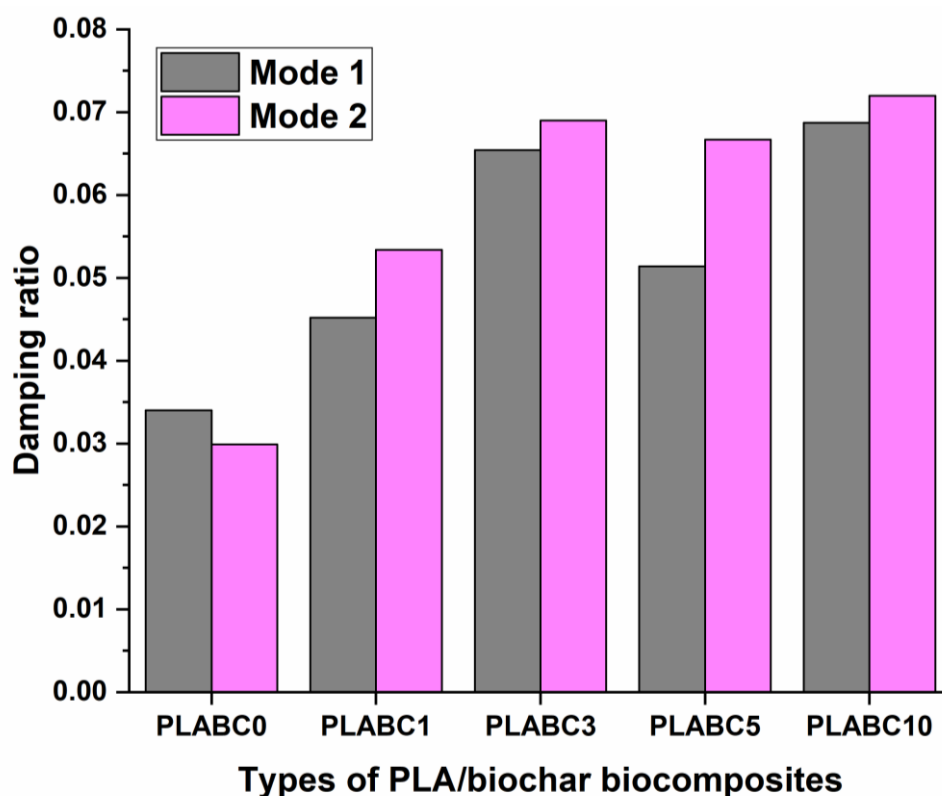


Figure 8 illustrates the impact of biochar filler on the damping ratio

Table 2 Description and composition of prepared PLA biocomposites via 3D printing

	Natural frequency	Damping ratio

PLA/biochar biocomposites	Mode 1	Mode 2	Mode 1	Mode 2
PLABC0	102	88	0.0340	0.0299
PLABC5	122	105	0.0422	0.0534
PLABC10	138	124	0.0514	0.0667
PLABC15	153	132	0.0654	0.0690
PLABC20	147	128	0.0687	0.0720

4. Conclusions

This study examines the effects of adding almond shell biochar particles on PLA biocomposites made using the FDM additive manufacturing technique. Following an analysis of the PLA biocomposites' mechanical and vibrational characteristics, the following results were made:

1. Biochar-filled PLA 3D printed biocomposites made from almond shells were successfully made using the FDM technique. Before 3D printing, the quality of the PLA extruded filament with 5% almond shell biochar was satisfactory. The filament quality was lower due to necking, discontinuity, and surface quality flaws when the amount of almond shell biochar was raised to roughly 10%. These flaws adversely impacted the mechanical qualities of biochar-filled 3D printed PLA biocomposites made from almond shells.
2. The interaction between the almond shell biochar particles and PLA matrix was greatly influenced by the distribution of the almond shell biochar particles, which changed the mechanical and vibration properties.
3. The PLA/almond shell biochar biocomposite's overall mechanical characteristics were improved by the 5% filler of biochar made from almond shells. The 5% PLA filled with almond shell biochar had tensile and flexural strengths of 61 MPa and 64 MPa, respectively.
4. By adding biochar up to 5 weight percent, the damped natural frequencies of biochar composites increased, and by adding more biochar, they dropped. When comparing PLA with biochar-filled composites to clean PLA composite, the largest improvements occurred at 5 wt%, reaching 49.01%. Compared to higher natural frequencies, the improvement in the fundamental natural frequency was more obvious. For the first mode of vibration, the damping ratios from biochar at 10 wt.% loading were higher

than those from all other biocomposites. A maximum improvement of 140.8% in damping ratio of biochar-filled biocomposite was observed at 10 wt.% loading.

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