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ORIGINAL RESEARCH

Rice husk/glass fiber-reinforced poly(lactic acid) hybrid composites: rheological and dynamic mechanical study

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Abstract
This study explores the effects of incorporating rice husk, glass fiber, and a POE-*g*-Mah(ethylene–octene copolymer functionalized with maleic anhydride) compatibilizer on the properties of poly(lactic acid) (PLA)-based composites. Through SEM analyses, it is observed that enhanced filler dispersion and interfacial bonding between the PLA matrix and fillers with POE-*g*-Mah result in improved composite compatibility. The rheological percolation threshold of the rice husk is determined above 30% for the samples containing only PLA and rice husk; however, it is determined between 20 and 30% for the samples containing the POE-*g*-Mah. The incorporation of POE-*g*-Mah into the polymer phase causes much higher modulus values compared to its counterparts with the same filler concentration. The DMA results revealed significant enhancements in the modulus of elasticity and damping properties when rice husk and glass fiber were added. The composite with 20% (by weight) rice husk and 5% (by weight) compatibilizer doubled its elastic modulus at 40 °C and increased fivefold to 65 °C with an additional 15% (by weight) glass fiber. This modification substantially reduced energy dissipation and improved damping performance and dimensional stability, which was also evidenced by a decrease in thermal expansion. These findings imply the potential of using rice husk, glass fiber, and POE-*g*-Mah to significantly enhance the mechanical and thermal properties, offering valuable insights for developing high-performance sustainable materials.

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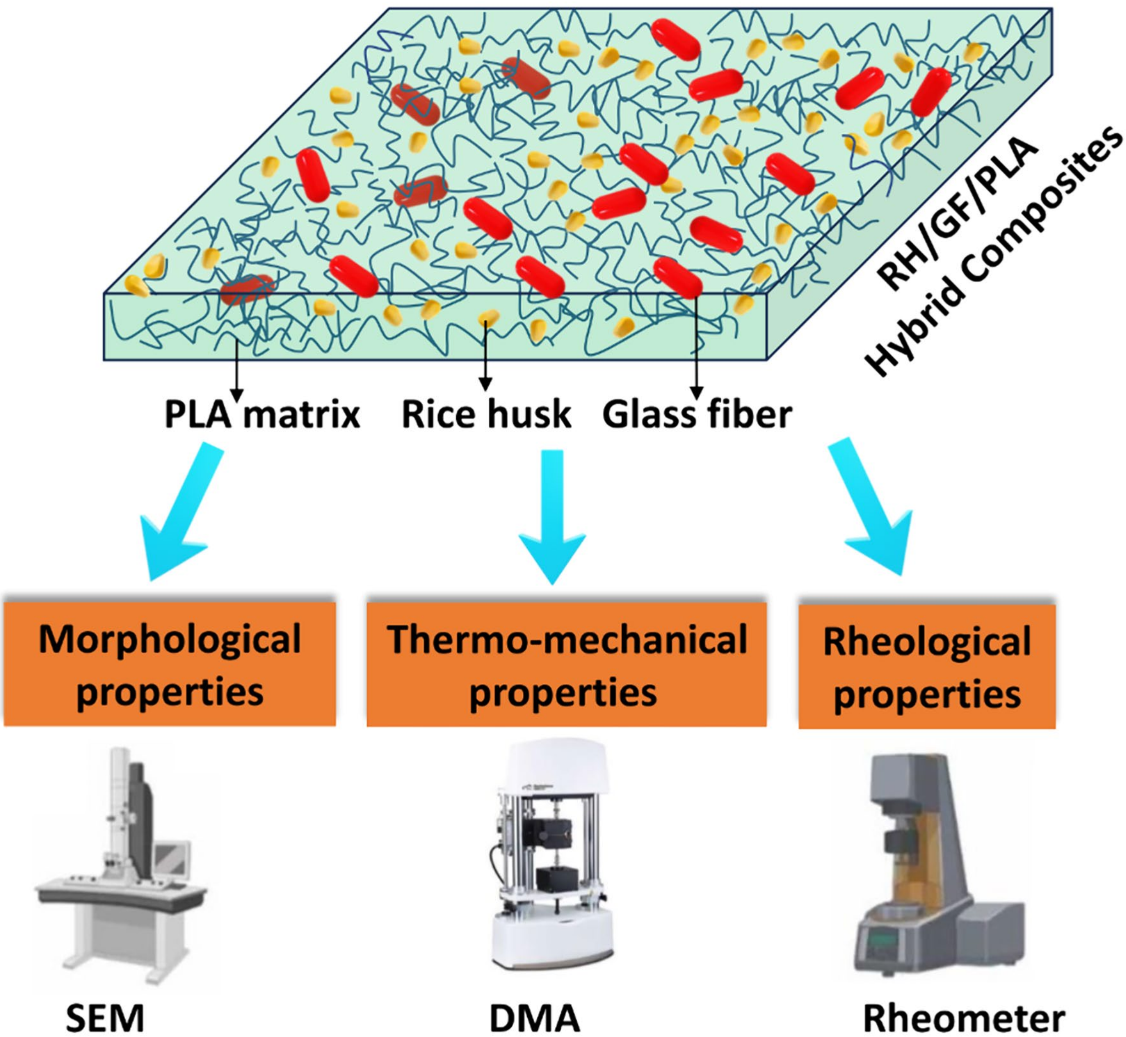
Graphical abstract



Keywords Glass fiber · Hybrid composites · Poly(lactic acid) · Rice husk · Thermoplastic

Introduction

The search for the ideal materials is constantly ongoing and material scientists try various combinations of reinforcements and processing methods to develop innovative materials for next-generation applications. As industries seek materials capable of withstanding superior mechanical properties while minimizing environmental impact, hybrid composites offer a promising shift in material design. It is made by combining biodegradable polymers with fibers and particles derived from natural sources to achieve a harmonious balance between strength, versatility, and eco-friendliness [1]. Poly(lactic acid) (PLA) is a promising bio-based polymer sourced from renewable materials like corn, starch, wheat straw, and sugarcane, among others [2]. Its eco-friendly features, affordability, biodegradability, sustainability, and reduced carbon footprint have made it a noteworthy substitute for conventional petroleum-derived plastics [3]. However, PLA has some limitations, including its brittleness, high permeability (moisture and oxygen pass more easily than other thermoplastics), and lower thermal stability, which can affect its overall performance in certain applications [4]. To overcome these limitations, the incorporation of reinforcing fillers such as synthetic (glass, carbon, aramid, and basalt) and natural (kenaf, jute, bamboo, cotton, and flax) fibers and particles (calcium carbonate, clay, mica, alumina, fly ash, silica, rice husk, hazelnut, walnut, pistachio and cocoa shells, wood sawdust, etc.) is a promising method to enhance the physical properties of PLA [5, 6]. Hybrid composites are materials made by combining two or more different types of reinforcing fillers into a single matrix [7, 8]. For example, combining glass fiber with graphene oxide can enhance the nucleation capability of PLA, significantly enhancing its crystallinity. As a result, the obtained hybrid composites exhibit better thermomechanical properties [9]. Similarly, the talc filler can induce the crystallization of PLA in the preparation of talc/natural fiber/PLA hybrid composites, leading to significant improvements in both tensile strength (by 11%) and elongation at break (by 30%) [10].

The rice husk is an agricultural by-product abundantly available globally, and it possesses unique properties such as low cost, low density, and high strength [11–13]. The rice husk acts as a filler, reinforcing the polymer matrix and enhancing mechanical properties such as tensile strength, flexural strength, and impact resistance [14]. Additionally, the incorporation of rice husk can lead to improvements in thermal stability, making the composites suitable for a broader range of applications [15–17]. The combination of PLA and rice husk results in composites that offer a synergistic effect, combining the biodegradability of PLA with the reinforcing and cost-effective attributes of rice husk. Utilizing rice husk as a filler in PLA composites not only addresses the issue of disposal of agricultural waste but also contributes to the development of sustainable materials with improved performance [18, 19].

In this study, the rice husk was used as a hybrid reinforcement in the strengthening of PLA/glassfiber composites. The effect of hybrid reinforcements either rice husk or glassfiber on the morphological, mechanical and rheological properties of the composites was systematically investigated. In addition, the low crystallinity ethylene–octene copolymer functionalized with maleic anhydride (POE-*g*-Mah) was utilized as a third component that functions as a compatibilizer between PLA and glass fiber and rice husk. These hybrid composites represent a promising avenue in the pursuit of sustainable materials with enhanced performance characteristics, offering potential applications in various industries such as packaging, automotive, and construction.

Experimental

Materials

PLA (Luminy L 130) was obtained from Luminy, Total Energies Corbion (Gorinchem, the Netherlands). Rice husk waste (particle size 16.59 µm) was supplied from a rice mill (Sakarya, Turkey). The low crystallinity ethylene–octene copolymer functionalized with maleic anhydride (POE-*g*-Mah, Paraloid™ EXL-3808) was supplied by Dow Chemical Company (Terneuzen, the Netherlands) and used as a compatibilizer. The crimped glass fiber (PA-2, E-type, nominal fiber diameter 11 µ, maximum moisture content 0.07%, silane (binder type), binder amount 0.60% ± 015%, polyamide (resin compatibility), and crop length 4.5 mm) was obtained from Sise Cam (Istanbul, Turkey).

Sample preparation

Rice husks and PLA were ground into fine powder using a miniature mechanical grinder (Renas Machines, Turkey, 2 kg capacity, 3000 rpm). The ground rice husk and PLA were sieved to an average particle size of 100 and 500 µm, respectively. The rice husk and PLA powders were dried under vacuum at 80 °C for 24 h before the extrusion process. A high shear force twin-screw co-rotating extruder (Gulnar Machines, Turkey, screw diameter 16 mm, L/D = 40) was used to produce the granules described in Table 1. A temperature profile inside the extruder barrel comprising between 140, 150, 165 and 170 °C was applied throughout the barrel from the feeding zone to the die.

Characterization

The SEM imaging was carried out by Quanta FEG 250 (Thermo Fisher Scientific, Hillsboro, OR, USA) instrument operating at an acceleration voltage of 15 kV. The

Table 1 Sample notations and compositions

Sample name	Filler type 1	Filler concentration 1 (% by weight)	Filler type 2	Filler concentration 2 (% by weight)
PLA	–	–	–	–
PLA-10rh	Rice husk	10		
PLA-20rh	Rice husk	20		
PLA-30rh	Rice husk	30		
PLAc*	–	–	–	–
PLAc-10rh*	Rice husk	10		
PLAc-20rh*	Rice husk	20		
PLAc-30rh*	Rice husk	30		
PLAc-20rh-5gf*	Rice husk	20	Glass fiber	5
PLAc-20rh-10gf*	Rice husk	20	Glass fiber	10
PLAc-20rh-15gf*	Rice husk	20	Glass fiber	15

*Polymer phase contains 5% (by weight) POE-*g*-Mah as compatibilizer surfaces of the samples were coated with gold to make the surface conductive before the SEM imaging. The rheological characteristics of the samples were examined using a rotational dynamic oscillatory rheometer, which features parallel plate geometry with a diameter of 25 mm (Discovery Hybrid Rheometer-1, DHR-1, TA, USA). Prior to assessing the rheological properties of the melt state of the samples through rheological measurements, an initial time sweep test was performed on the neat PLA sample. This test, conducted at 200 °C with strain amplitude of 1% and a frequency of 1 rad/s, aimed to determine the thermal stability of the polymer phase during rheological testing. Following this, a dynamic strain sweep test was performed on the samples, employing a strain range from 0.1% to 10% and an angular frequency of 10 rad/s, to identify their linear viscoelastic region. Subsequently, frequency sweep tests were applied over a frequency range of 100 to 0.1 rad/s at a fixed strain of 1%. Additionally, to evaluate the shear viscosity of the samples, a steady-state flow test was conducted at 200 °C, within a shear rate spectrum 0.1–100 s^{-1}. Dynamic mechanical analysis (DMA) and thermal expansion measurements of the samples were conducted using a discovery hybrid rheometer (Discovery Hybrid Rheometer-1, DHR-1, TA, USA) fitted with a solid-state tension test configuration. An oscillatory force was applied at a frequency of 1.0 Hz across a temperature range of 30–75 °C, accompanied by a constant low-level stress (0.1 MPa) and a heating rate of 1 °C/min.

Results and discussion

Morphological properties

To investigate the effects of compatibilizer, glass fiber, and rice husk addition on the microstructural properties of the hybrid composite, such as filler dispersion and filler/filler or filler/polymer interactions, SEM analysis was conducted on composites with different formulations, all maintaining constant rice husk concentrations. The SEM images of the samples containing 20% (by weight) rice husk (PLA-20rh, PLAc-20rh and PLAc-20rh-10gf) are presented in Fig. 1. In addition, the energy-dispersive X-ray (EDX) spectra of the samples confirmed the presence of silica and aluminum

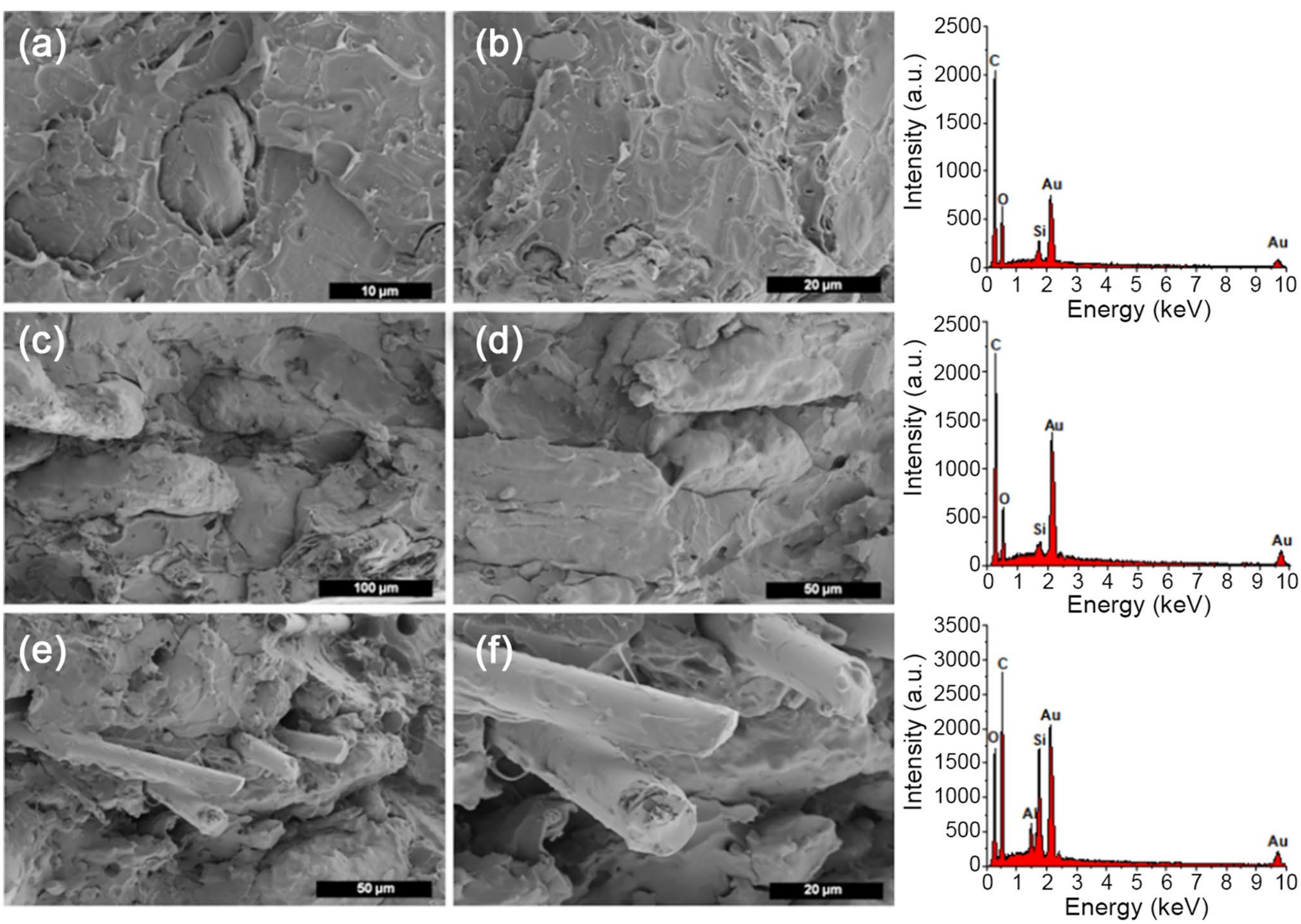


Fig. 1 SEM images and EDS graphics: (**a**–**b**) PLA-20rh, (**c**–**d**) PLAc-20rh, and (**e**–**f**) PLAc-20rh-10gf at different magnifications

elements, which are the fundamental components of rice husk and glass fiber, respectively.

Figure 1a, b of particle size of the rice husk varied between 5 and 10 µm throughout the PLA-20rh sample. Additionally, the gaps between rice husk and PLA phase illustrate the poor interfaces. On the other hand, the SEM images of the PLAc-20rh sample in Fig. 1c, d shows that the rice husk particles were entirely covered with the PLA matrix, indicating that the compatibility between the rice husk particles, and the polymer phase was significantly improved through the addition of the compatibilizer. The SEM micrographs of PLAc-20rh-10gf in Fig. 1e, f demonstrate that the compatibilizer also has a positive effect on greatly enhancing the coating of both glass fiber and rice husk particles within the PLA phase. However, especially in regions where glass fiber agglomerates existed, gaps between the PLA phase and the glass fibers were visible. These gaps, which were observed despite the positive effect of the compatibilizer, confirmed that the structure of the compatibilizer was suitable for the preparation of rice husk/glass fiber-reinforced PLA hybrid composites. On the other hand, a higher amount of compatibilizer in the composite structure appeared to provide a more effectively compatible composite structure for the composite containing glass fiber and rice husk simultaneously.

Rheological properties

Prior to conducting rheological analyses, a time sweep test was performed on the neat PLA to assess any thermal degradation that might influence the polymer phase rheological characteristics during the measurements. In this test, variations in the viscoelastic parameters of the sample are followed with time under constant test parameters (angular frequency, shear strain, and temperature). This test is also widely used to determine the maximum residence time of the polymer phase in the melt processing conditions. In this test, the changes in G' and G'' values over time were followed for 30 min, and the obtained experimental values are given in Fig. 2.

Figure 2 shows that the G' and G'' values of the neat PLA does not significantly change over 30 min. Therefore, it may be concluded that PLA does not undergo any considerable structural decomposition when exposed to a temperature of 190 °C for 30 min. In this context, the maximum test time was established by 30 min for all samples in melt state rheological characterization. The G' (storage modulus) and G'' (loss modulus) values of the samples, which depend on angular frequency, were determined using the frequency sweep test (Fig. 3).

As anticipated, both modulus values exhibit a decrease with the reducing angular frequency due to a reduced deformation rate, as expected. However, higher modulus values

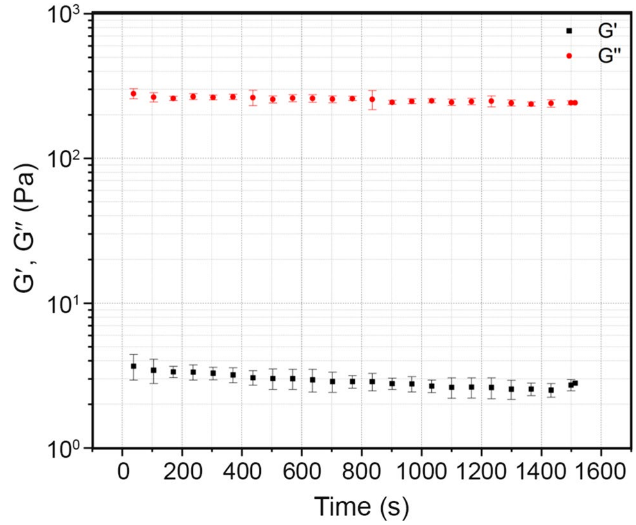


Fig. 2 Variations in G' and G'' values of PLA with time

were measured in the high-frequency region for all samples, and this can be explained by the viscoelastic structure of the sample approaching rigid (elastic) behavior due to the shorter time allowed for the chains to rearrange after each oscillation deformation. Figure 3a, b shows that the addition of rice husk by 10% into the PLA phase caused a significant increase in the G' and G'' values in the entire angular frequency region. Additionally, it was observed that increasing the rice husk concentration beyond 10% did not yield a substantial rise in the modulus values within the high-frequency domain. Nevertheless, a slight increase in the modulus values with increasing rice husk concentration could be obtained without changing the decreasing trend depending on the angular frequency in the low-frequency region. On the other hand, Fig. 3 shows that the angular frequency-independent modulus values, which are defined as solid-like behavior in composite rheology and occur in low-frequency regions due to the physical interaction of the solid filler (percolation), could not be obtained [20, 21]. Thus, it was evaluated that the percolation threshold of the rice husk was above 30% in the sample series, which contained only PLA and rice husk. The G' and G'' values of the samples containing 5% compatibilizer and the same amount of rice husk are presented in Fig. 3c, d. The comparison of Fig. 3a, b with Fig. 3c, d indicated that the incorporation of the compatibilizer into the polymer phase caused much higher modulus values. The increase in the modulus values was associated with an improved interphase interaction between the rice husk and PLA phase, confirmed by previous SEM observations. Another remarkable point was the frequency-independent G' values in the terminal region, which are not observed in Fig. 3a, b. Firstly, the addition of the compatibilizer increased the interaction between the

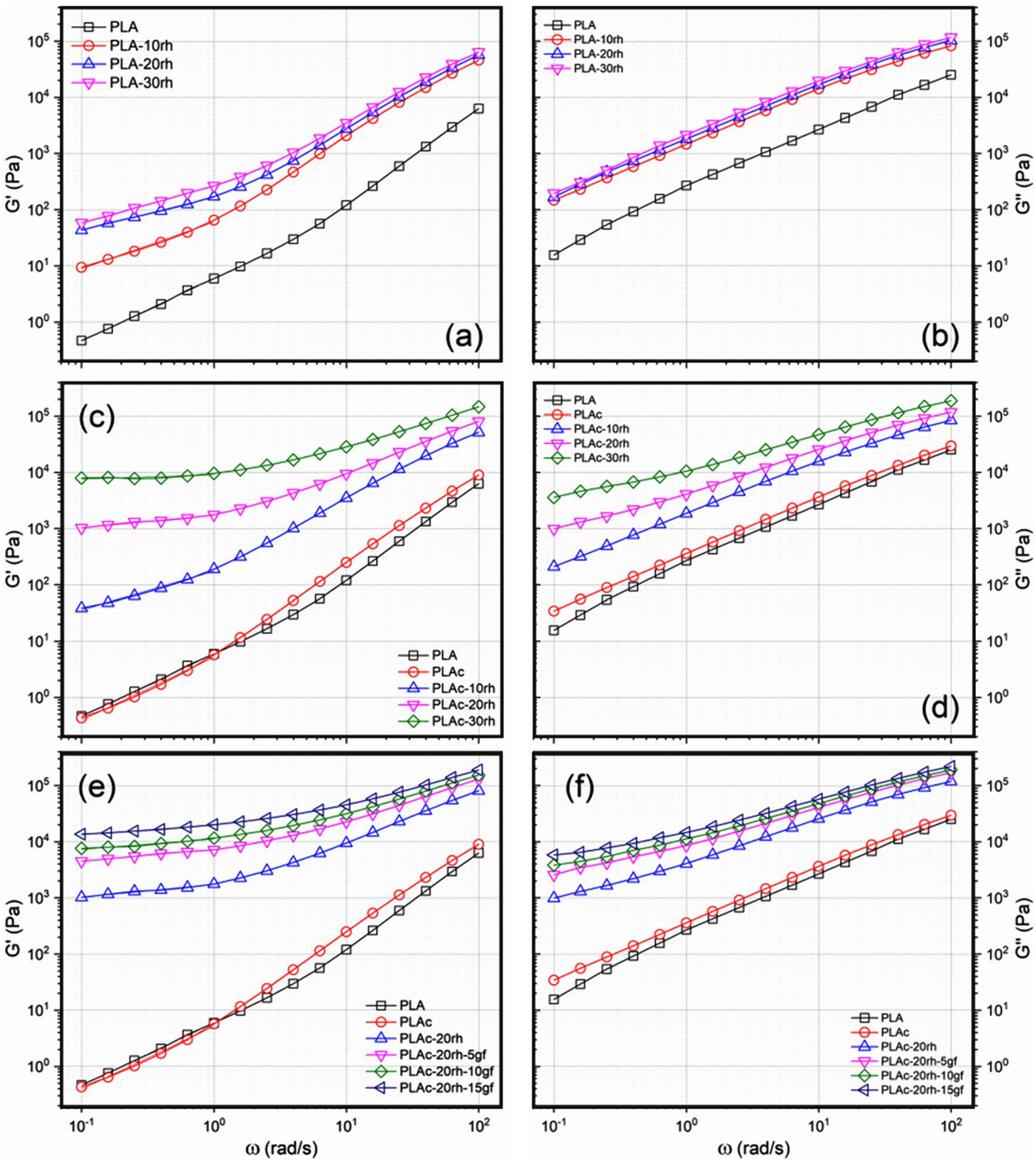


Fig. 3 Variations in G' and G'' values of the obtained samples with angular frequency

rice husk and the PLA phase, thus reducing the slippage at filler/polymer interfaces. With the increase in the rice husk concentration, especially in the low-frequency region, the influence of the filler phase on the modulus values increased and started to determine the rheological behavior of the composite. This critical filler concentration at which the filler phase determines the overall rheological behavior of the composite (the G' parameter takes a constant value independent of the frequency) is defined as the rheological percolation threshold. From this point of view, it can be concluded that the rheological percolation of the filler occurs between 20 and 30% for the PLAc series of samples. In Fig. 3e, f, it is observed that the incorporation of glass fiber into the PLAc samples, which contain 20% (by weight) of rice husk, enhances the modulus values by more than tenfold. However, this addition does not seem to alter the relationship between the modulus values and the angular frequency. Furthermore, Fig. 3a–f demonstrates that the addition of both fillers and compatibilizers to PLA significantly improved its elastic properties (G') in the melt state more than its viscous properties (G'')."

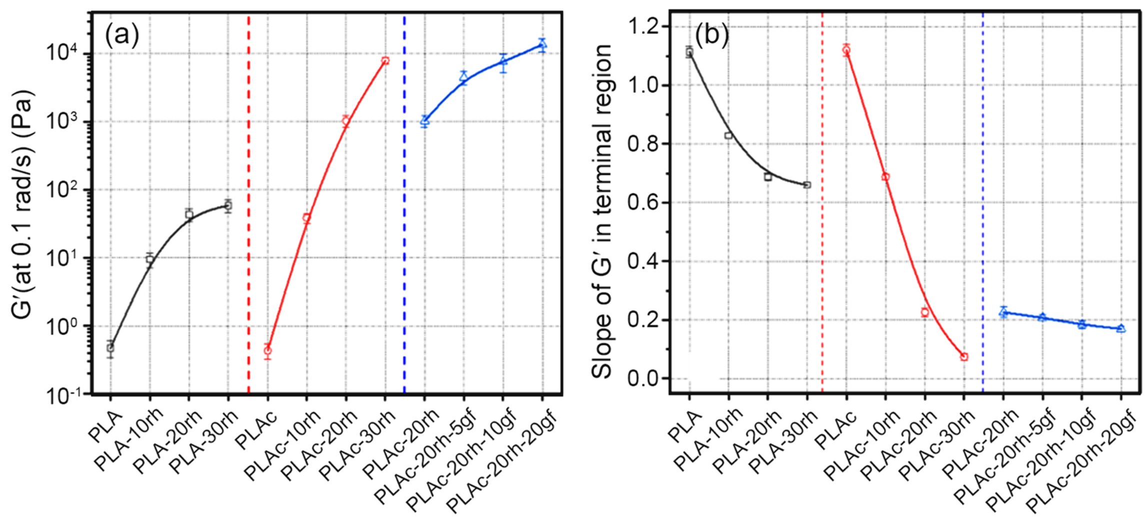


Fig. 4 **a** G' values of the samples at 0.1 rad/s, and **b** slope of G'-ω line in the terminal region

To investigate how sample composition affects the variation in G' values, the modulus values at the lowest frequency (0.1 rad/s) are calculated and displayed in Fig. 4a. Additionally, the slope of the G'-ω curves across the angular frequency range of 1–0.1 rad/s is provided in Fig. 4b. Figure 4a reveals that the improvement in modulus values is less evident with increasing filler concentration, particularly for PLA series samples with a filler concentration of 30%. Additionally, it is observed that the G' values of the PLA-30 sample are close to those of the PLAc-10 sample, and their modulus value is increased approximately 100 times with the addition of rice husk in the PLAc sample series. It is also noteworthy that the PLAc-20-10gf and PLAc-30 samples, despite containing the same amount of filler, exhibit similar modulus values. In addition, it was predicted that the addition of glass fiber would increase the modulus values, but the expected increase was not achieved. This deficiency has been attributed to the inability to achieve effective interphase adhesion between fillers and PLA phase as a result of keeping the compatibilizer at a constant concentration. The slope of the G'–ω plots in the terminal region in Fig. 4b shows an almost frequency-independent modulus behavior for a PLAc-30rh sample, while higher slope values are observed for the samples containing the glass fiber. The lower slope values of PLAc-30rh, which contains the same weight of filler as PLAc-20rh-10gf, compared to PLAc-20-10gf, may indicate that the compatibilizer forms a better interaction with the rice husk compared with the glass fiber.

To examine the rheological properties of the prepared composites during melt processing, the variations in their shear viscosity values, as the function of shear rate, were examined, and flow curves were modeled using the Cross model. The experimentally obtained flow curves are presented in Fig. 5. In Fig. 5a, it is observed that all samples without compatibilizers exhibit Newtonian characteristics in the shear rate range of 0.1–10 s^{-1} and shear-thinning behavior above a shear rate of 10 s^{-1}.

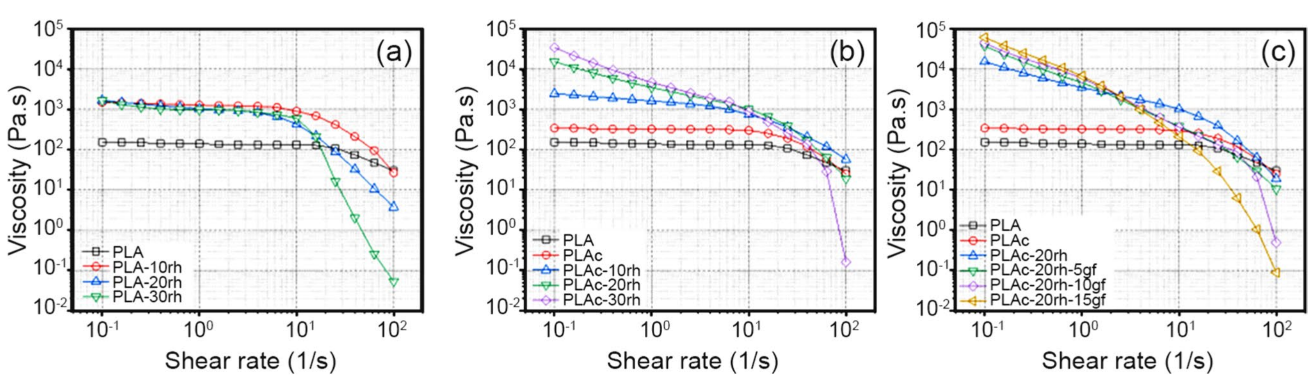


Fig. 5 Shear rate-dependent shear viscosity values of the samples

In the shear-thinning region, concerning the orientation of the glass fiber particles, it was observed that the viscosity values of the glass fiber-reinforced composites with higher filler concentrations decreased more significantly compared to the samples with lower filler concentrations. As a result, they exhibited lower viscosity values than the unfilled PLA phase at the higher shear rate values. In Fig. 5b, it is observed that samples containing both compatibilizer and rice husk at concentrations exceeding 10% demonstrated shear-thinning characteristics across the entire shear rate range; however, this behavior was more pronounced at the higher shear rate of 10 s^{-1}, which was similar to that observed in samples without compatibilizer. Finally, Fig. 5c shows that the addition of glass fiber resulted in increased viscosity values at shear rates below the critical shear rate (approximately 10 s^{-1}), while it led to a drop in viscosity at higher rates.

The mathematical modeling of flow curves for polymer materials is a widely used method to determine the behavior of polymers in all melt molding methods, especially extrusion and injection molding. One such model is the Cross model [21], the mathematical expression of which is provided in Eq. 1. The parameters obtained by applying the Cross model to the flow curves are presented in Table 2 [22].

$$\frac{\eta - \eta_\infty}{\eta_0 - \eta_\infty} = \frac{1}{1 + (K\gamma)^{m}}. \tag{1}$$

In Eq. 1, η_0 represents the zero-shear rate viscosity, while K and m indicate the consistency and the rate index, respectively. Additionally, η_0 is primarily utilized to determine the behavior of the properties related to molecular structure, such as molecular weight, as well as the behavior of polymers in melt process techniques with very low shear rates, for example, compression molding.

The comparison of η_0 values in Table 2 shows that, as expected, η_0 is increased with the addition of rice husk. It should be noted that these increases were achieved more effectively with the addition of a compatibilizer. Furthermore, it was observed that the glass fiber caused a greater increase in η_0 values, in contrast to the viscosity values at high shear rate values. The critical shear rate (γ_c), indicating the transition of a polymer melt rheological behavior from Newtonian to shear thinning, is determined by the equation as 1/K in s^{-1}. Table 2 shows a decrease in γ_c as filler concentrations rise. Changes in the power-law coefficient may imply the way the viscosity of the samples responds to varying shear rates. It is noted that the shear sensitivity of the samples is increased with higher filler content, a trend that is particularly evident in the series of glass fiber-reinforced samples.

Table 2 Cross model parameters of the samples

Samples	η_0 (Pa.s)	m	K	R^2
PLA	140.6	2.183	0.0264	0.993
PLA-10rh	1361.8	1.605	0.0632	0.998
PLA-20rh	1221.3	2.046	0.1414	0.995
PLA-30rh	1029.6	4.948	0.0883	0.997
PLAc	326.5	1.960	0.0330	0.999
PLAc-10rh	2253.1	0.857	0.2111	0.997
PLAc-20rh	39,006.1	0.651	26.864	0.992
PLAc-30rh	6.377E7	0.724	406,389	0.997
PLAc-20rh-5gf	109,206	1.048	20.999	0.999
PLAc-20rh-10gf	94,384.4	1.100	12.900	0.998
PLAc-20rh-15gf	37,221.2	1.919	1.8323	0.991

Dynamic mechanical analyses (DMA)

Dynamic mechanical analysis (DMA) is a method used to examine the thermo-mechanical and viscoelastic (E', E'' and tanδ) properties of solid-state samples over a specific temperature range. It is also regarded as a characterization technique for polymers because it allows the determination

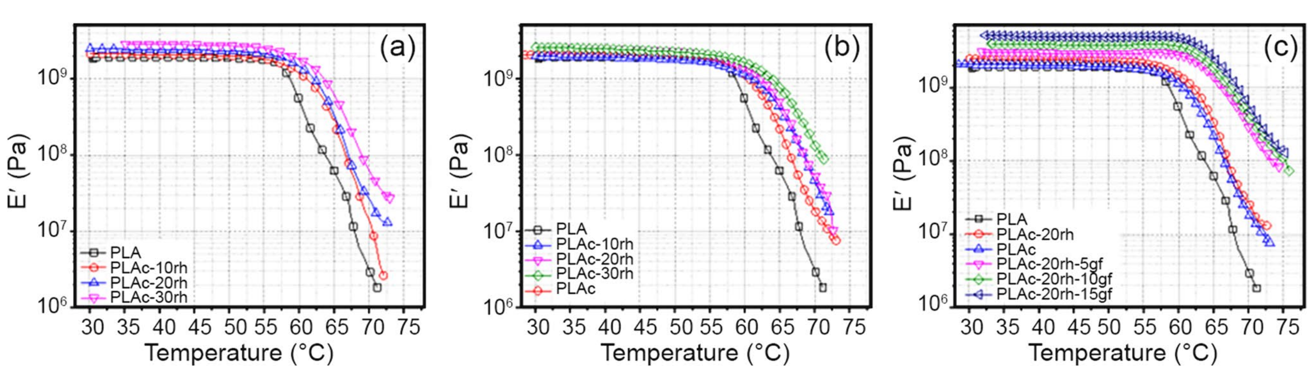


Fig. 6 Variations in E' values of the samples with temperature

of thermal transition temperatures, including melting, glass transition (T_g), and α-relaxation temperatures. The E' values of the samples for the temperature range of 35–75 °C are given in Fig. 6. As shown in Fig. 6, there is no significant change in the modulus values of all samples toward the T_g of PLA, which is approximately 60 °C. Above this temperature, as expected, the modulus values are decreased rapidly. The decrease in the E' values at the T_g is attributed to the increased mobility of the polymer chains with increasing temperature. In this regard, the decrease in modulus values at similar temperatures across all composite samples indicates that the incorporation of fillers or compatibilizers does not significantly alter the T_g of PLA.

Figure 6a, b demonstrates that adding rice husk to the PLA phase does not result in a significant increase in modulus values within the temperature range below the T_g, known as the glassy region. However, noticeable increases in the E' values with filler concentration are observed at temperatures above the T_g. Many previous studies have also indicated that the reinforcing capacity of fillers in amorphous polymers is much higher at the temperature range above the T_g of polymer phase [23–25]. Moreover, it has been observed that the addition of glass fiber to the composite structure leads to much more significant changes in modulus values in both the glassy and rubbery regions. The impact of adding rice husk, compatibilizer, and glass fiber to the polymer phase on modulus values was examined in more detail, and the results for the samples for two different temperatures (40 and 65 °C) are summarized in Fig. 7. In this case, the addition of a compatibilizer did not have a significant effect on the improvement of the solid-state modulus of elasticity values of the composites containing only rice husk. Furthermore, an exponential increase in modulus values was observed with the incorporation of glass fiber into the composite at both temperatures. It was also noted that the E' values of the sample containing 20% (by weight) rice husk and 5% (by weight) compatibilizer (PLAc-20rh) doubled at 40 °C and increased fivefold at 65 °C with the addition of 15% (by weight) glass fiber.

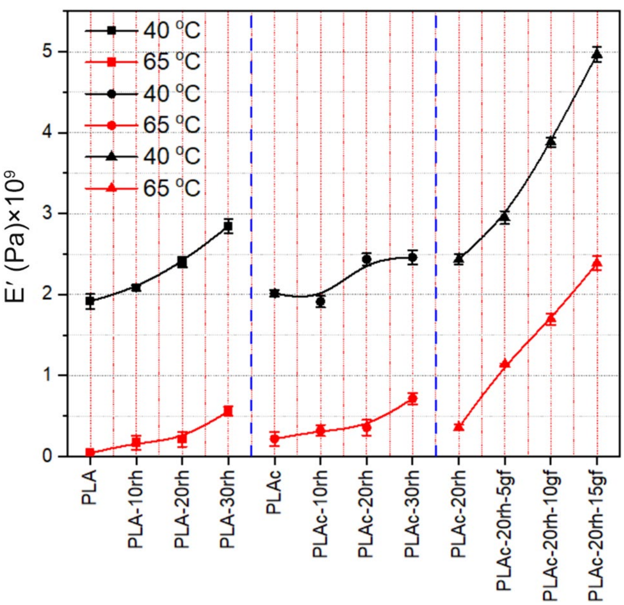


Fig. 7 Variations in E' values with composition

In Fig. 8, the variations of tanδ, which is an indicator of the energy dissipation capabilities of the samples, are presented as a function of temperature. The changes in the tanδ values showed significant differences between the samples around 60 °C, although no significant change was observed below their T_g, which corresponds to the service temperature of PLA. A decrease in the peak height of tanδ plots is interpreted as an increase in energy absorption capability [26]. In this context, the peak heights in Fig. 8a demonstrate that the addition of rice husk enhances the impact damping properties of PLA, while Fig. 8b shows that the impact damping performance increases more effectively when the compatibilizer is additionally added in the polymer phase. The lower tanδ peak heights in the samples containing both compatibilizer and rice husk, compared to

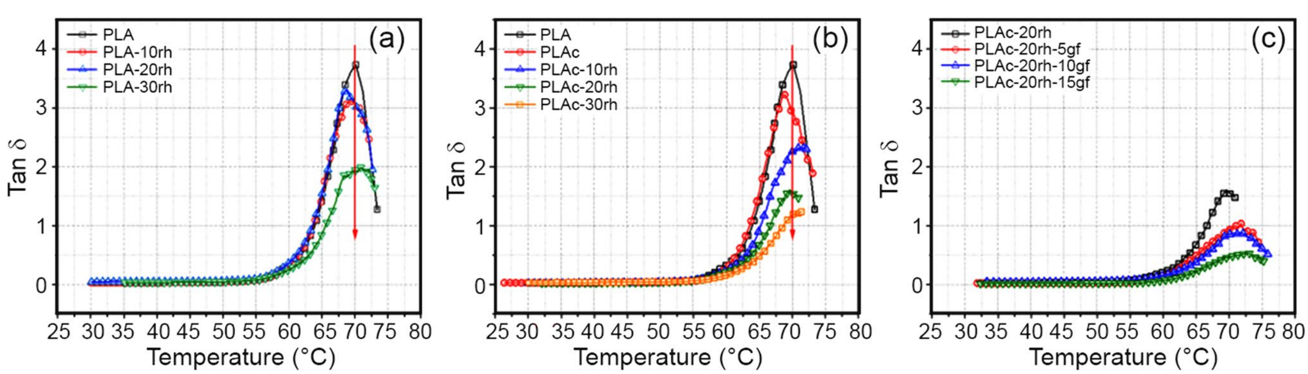


Fig. 8 Variations in tanδ values of the samples with temperature

those without compatibilizer, suggest that the compatibilizer has effectively contributed to the solid phase and successfully enhanced the structural compatibility of the samples [27]. Figure 8c reveals that the addition of glass fiber has reduced the energy dissipation performance more effectively. The tanδ peak height of composites reinforced with 15% glass fiber is lowered to about a seventh to eighth of the initial value compared to neat PLA, indicating a significant improvement in damping ability.

Thermal expansion coefficient

One of the critical parameters for PLA-based composites, particularly in 3D printing and injection molding, is dimensional stability. The dimensional stability of materials can be analyzed under two categories: the dimensional change (thermal expansion coefficient) that occurs with an increase in temperature under very low tension, and the change over time (creep) that happens under constant temperature and tension. For a polymer with a service temperature below its T_g, it is preferable to examine the thermal expansion coefficient instead of creep behavior due to the limited chain mobility of the polymer.

In this context, dimensional changes in the PLA, PLAc-20rh, PLA-20rh, and PLAc-20rh-10gf samples with increasing temperature were analyzed using the TMA method to assess the effects of different components on the thermal expansion coefficient, and the results are presented in Fig. 9a. As expected, no significant dimensional change was observed until reaching the T_g of PLA, which is approximately 65 °C; however, a rapid dimensional change was noted above this temperature. It was also found that the addition of glass fiber particularly enhanced dimensional stability by reducing deformation. The thermal expansion coefficient (α) value is determined by the plot of displacement (dL) against temperature (T) using Eq. 2. While the thermal expansion coefficient of pure PLA was found to be approximately 27, it was observed that the thermal expansion coefficient of composites containing 20% (by weight) rice husk dropped to around 12.5–13 [28, 29]. On the other hand, with the addition of 10% (by weight) of glass fiber, the coefficient of thermal expansion was reduced to 8.75, indicating an improvement of approximately 68% in dimensional stability compared to pure PLA.

$$a\left(K^{-1}\right) = \frac{dL}{L}\frac{1}{T} \tag{2}$$

Conclusion

The influence of incorporating rice husk, glass fiber, and a compatibilizer (POE-*g*-Mah) on the properties of PLA-based hybrid composites was comprehensively investigated by morphological, rheological, and thermo-mechanical analyses. Improved dispersion and compatibility of the rice husk filler was achieved with the addition of compatibilizer, which facilitated better interfacial interactions. Rheological assessments through time sweep, frequency sweep tests, and flow curve analysis revealed significant alterations in the viscoelastic properties and viscosity behaviors of the obtained composites. The inclusion of rice husk and compatibilizer enhanced the elastic properties more than their viscous properties of the composites, indicating a shift towards a more solid-like behavior. Based on DMA analysis results, the rice husk did not significantly affect the PLA glass transition

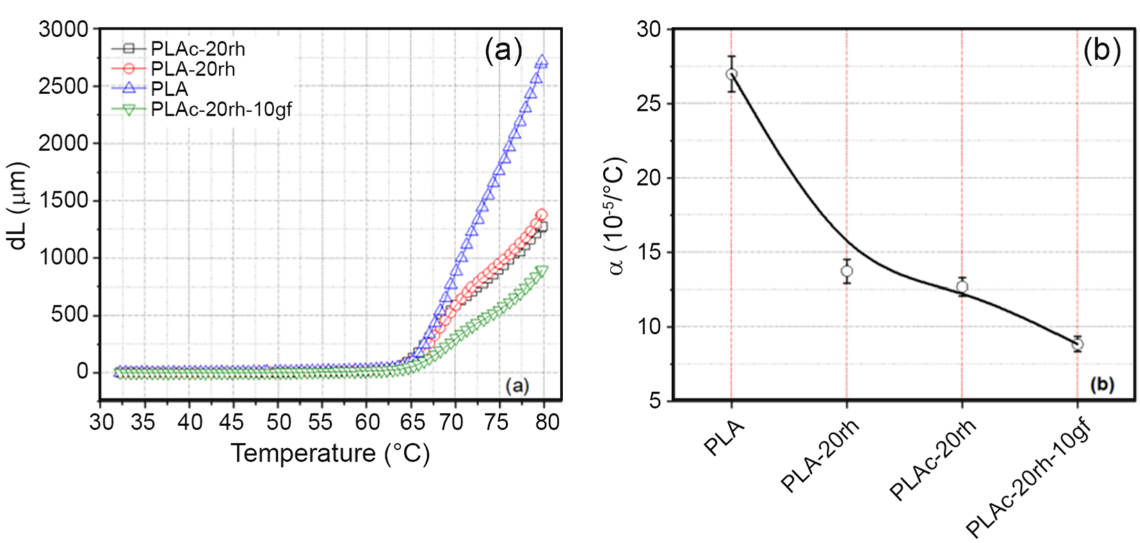


Fig. 9 **a** dL and **b** thermal expansion coefficient of the samples

temperature but improved its modulus of elasticity markedly, particularly above this temperature, and enhanced damping properties. Moreover, the addition of rice husk and glass fiber was found to significantly increase both the modulus values across the glassy and rubbery regions and improve dimensional stability by approximately 68%, as evidenced by a reduced thermal expansion.

By combining morphological, rheological, and thermomechanical analyses, this study illustrates the complex interactions between rice husk addition and the resultant properties of PLA hybrid composites. These findings offer critical insights into designing and manufacturing PLA-based materials with tailored functionalities, emphasizing the potential of rice husk, compatibilizers, and glass fiber in enhancing the performance characteristics of PLA composites for specific applications.

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Data availability The authors declare that the data supporting the findings of this study are available within the paper. Should any raw data files be needed in another format they are available from the corresponding authors upon reasonable request.

References

1. Satyanarayana KG, Arizaga GGC, Wypych F (2009) Biodegradable composites based on lignocellulosic fibers: an overview. Prog Polym Sci 34:982–1021
2. Mohanty A, Ma M, Hinrichsen G (2000) Biofibres, biodegradable polymers and biocomposites: an overview. Macromol Mater Eng 276:1–24
3. Pang X, Zhuang X, Tang Z, Chen X (2010) Polylactic acid (PLA): research, development and industrialization. Biotechnol J 5:1125–1136
4. Inkinen S, Hakkarainen M, Albertsson AC, Södergård A (2011) From lactic acid to poly (lactic acid) (PLA): characterization and analysis of pla and its precursors. Biomacromol 12:523–532
5. Oksman K, Skrifvars M, Selin JF (2003) Natural fibres as reinforcement in polylactic acid (PLA) composites. Compos Sci Technol 63:1317–1324
6. Rajeshkumar G, Seshadri SA, Devnani GL, Sanjay MR, Siengchin S, Maran JP, Al-Dhabi NA, Karuppiah P, Mariadhas VA, Sivarajasekar N (2021) Environment friendly, renewable and sustainable poly lactic acid (PLA) based natural fiber reinforced composites: a comprehensive review. J Cleaner Prod 310:127483
7. Safri SNA, Sultan MTH, Jawaid M, Jayakrishna K (2018) Impact behaviour of hybrid composites for structural applications: a review. Compos B 133:112–121
8. Singh J, Kumar M, Kumar S, Mohapatra SK (2017) Properties of glass-fiber hybrid composites: a review. Polym Plast Technol Eng 56:455–469
9. Jing M, Che J, Xu S, Liu Z, Fu Q (2018) The effect of surface modification of glass fiber on the performance of poly (lactic acid) composites: Graphene oxide vs. Silane Coupling Agents Appl Surf Sci 435:1046–1056
10. Nanthananon P, Seadan M, Pivsa-Art S, Hamada H, Suttiruengwong S (2018) Facile preparation and characterization of short-fiber and talc reinforced poly (lactic acid) hybrid composite with in situ reactive compatibilizers. Materials 11:1183
11. Halip JA, Lee SH, Tahir PM, Chuan LT, Selimin MA, Saffian HA (2021) A review: chemical treatments of rice husk for polymer composites. Biointerface ResAppl Chem 11:12425–12433
12. Am Aridi N, Sapuan M, S, S Zainudin E, M Al-Oqla F, (2017) A review of rice husk bio-based composites. Curr Org Synth 14:263–271
13. Suhot MA, Hassan MZ, Aziz SaA Md, Daud MY (2021) Recent progress of rice husk reinforced polymer composites: a review. Polymers 13:2391
14. Aa Z, Zanganeh A, Hemmati F, Mohammadi-Roshandeh J (2019) Thermal and biodegradation properties of poly (lactic acid)/rice straw composites: effects of modified pulping products. Iran Polym J 28:403–415
15. Ghorbani Chaboki M, Mohammadi-Rovshandeh J, Hemmati F (2019) Poly (lactic acid)/thermoplasticized rice straw biocomposites: effects of benzylated lignocellulosic filler and nanoclay. Iran Polym J 28:777–788
16. Yee YY, Gunathilake T, Ching YC (2023) PLA-based composite film reinforced with empty fruit bunch fiber and silica for food packaging applications. Iran Polym J 32:1367–1376
17. Adamu MA, Sumaila M, Dauda M, Ause T (2024) A novel polycaprolactone/rice husk ash/hydroxyapatite biopolymer composite for bone implant: physico-mechanical and biodegradable analysis. Iran Polym J 33:395–404
18. Nizamuddin S, Jadhav A, Qureshi SS, Baloch HA, Siddiqui MTH, Mubarak NM, Griffin G, Madapusi S, Tanksale A, Ahamed MI (2019) Synthesis and characterization of polylactide/rice husk hydrochar composite. Sci Rep 9:5445
19. Wu CS, Tsou CH (2019) Fabrication, characterization, and application of biocomposites from poly (lactic acid) with renewable rice husk as reinforcement. J Polym Res 26:1–9
20. Penu C, Hu GH, Fernandez A, Marchal P, Choplin L (2012) Rheological and electrical percolation thresholds of carbon nanotube/polymer nanocomposites. Polym Eng Sci 52:2173–2181
21. Hassanabadi HM, Wilhelm M, Rodrigue D (2014) A rheological criterion to determine the percolation threshold in polymer nanocomposites. Rheol Acta 53:869–882
22. Rudolph NM, Agudelo AC, Granada JC, Park HE, Osswald TA (2016) WLF model for the pressure dependence of zero shear viscosity of polycarbonate. Rheol Acta 55:673–681
23. Droste DH, Dibenedetto AT (1969) The glass transition temperature of filled polymers and its effect on their physical properties. J Appl Polym Sci 13:2149–2168
24. Kasgoz A, Akın D, Ayten AI, Durmus A (2014) Effect of different types of carbon fillers on mechanical and rheological properties of cyclic olefin copolymer (COC) composites. Compos B 66:126–135
25. Akın D, Kasgoz A, Durmus A (2014) Quantifying microstructure, electrical and mechanical properties of carbon fiber and expanded graphite filled cyclic olefin copolymer composites. Compos A 60:44–51
26. Mohamed SAN, Zainudin ES, Sapuan SM, Azaman MD, Arifin AMT (2020) Energy behavior assessment of rice husk fibres reinforced polymer composite. J Mater Res Technol 9:383–393
27. Haris NIN, Hassan MZ, Ilyas RA (2022) Crystallinity, chemical, thermal, and dynamic mechanical properties of rice husk/coco peat fiber reinforced abs biocomposites. J Nat Fibers 19:13753–13764

28. Fuad MYA, Mustafah J, Mansor MS, Ishak ZAM, Omar AKM (1995) Thermal properties of polypropylene/rice husk ash composites. Polym Int 38:33–43
29. Halim ZAA, Yajid MAM, Idris MH, Hamdan H (2018) Effects of rice husk derived amorphous silica on the thermal-mechanical properties of unsaturated polyester composites. J Macromol Sci Part B Phys 57:479–496