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Influence of $BaSO_4$ Microfiller on Mechanical, Dynamic, and Interfacial Behavior of Banana/Sisal Hybrid Epoxy Composites for Automobile Application

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This study focused on the fabrication of composite materials by combining banana and sisal fibers and adding fillers, such as epoxy resin and Barium Sulfate (BS). The objective of this study was to investigate the impact of various filler weight percentages on the material strength and flexibility. The composites were fabricated with different weight percentages using the hand lay-up method, and mechanical and flexural tests were conducted in accordance with the ASTM standard. Additionally, field emission scanning electron microscopy of the broken surface of the composite was performed to determine the amount of fiber–matrix interaction and filler dispersion and to identify stability. Water absorption tests were conducted to evaluate environmental stability. The results suggested that the addition of BS resulted in increased load transfer and stiffness rather than strength, and the composites with moderate filler contents displayed superior flexural behavior and stronger impact resistance. Furthermore, higher filler contents produced particle agglomeration and, thus, the creation of microvoids, which led to worse performance. It was also observed that there was a gradual increase in the mechanical properties when fillers were added up to 6 wt%. The TS increased by 23.21%, and the FS increased by 18.91%. Finally, the addition of fillers to the composite made from banana and sisal fibers produced better properties and can be used as biodegradable materials.

Keywords: Hybrid Composites, natural microfiller, mechanical and morphological properties, structural applications

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The increasing importance of sustainable materials and the progressive exhaustion of existing resources have driven the development of natural fiber-reinforced polymer composites in the engineering industry. These materials are characterized by low density, appropriate strengths, and lower environmental effects compared to glass and carbon fiber composites [1, 2]. However, they cannot always perform well in real-life applications. Natural fibers are absorbent, and the absorption of moisture usually causes dimensional instability, poor interfacial bonding, and gradual loss of mechanical strength. These limits limit their direct applicability in structural elements [3, 4]. Recent studies have focused on strengthening the fiber–matrix interaction via hybridization and the addition of fillers instead of depending on a single-fiber system [5, 6]. Combining diverse natural fibers can assist balance stiffness, flexibility, and energy absorption. Banana and sisal fibers are typically mentioned in the same context since they differ in terms of mechanical reaction but can still be employed in processing [7]. Banana fibers are not overly stiff and contribute toughness, whereas sisal fibers make them more rigid and offer tensile protection [8–9]. Once these fibers are incorporated in an epoxy matrix, a more stable weight distribution is achievable, and the risk of unexpected failure is minimized. Hybrid banana–sisal laminates have been proved to be more effective than single-fiber composites in tensile and flexural loading, however the differences significantly depend on the quality of fabrication and exposure to the environment [10, 11]. Although hybridization has been achieved, the challenges of moisture uptake and interfacial compatibility remain unresolved,

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especially when the matrix fails to supply appropriate microstructural support [12].

Inorganic fillers have consequently been employed to fill polymer systems to overcome these difficulties and fortify the matrix and stabilize the interface. Barium sulfate – $BaSO_4$ (BS) is one such filler that has been exploited because of its thermal stability, chemical inertness, and ability to combine with epoxy resins. In its dispersed condition in the matrix, it may inhibit the movement of polymer chains and increase the hardness and stiffness [13-14]. It can also limit the channels of moisture diffusion, which is significant for natural fiber composites under humid conditions [15-16]. Nevertheless, the effectiveness of BS is not straightforward. When the particles are not well-dispersed, they gather together, generating localized stress regimes that may produce cracks. Filler content beyond a specific point does not necessarily result in further enhancement and can, in fact, result in worse performance [17]. Previous studies on hybrid natural fiber composites have also examined various combinations of fibers, stacking patterns, and surface finishing to enhance the behavior of the material in terms of mechanical behavior [18]. There are studies that have indicated increased damping and viscoelastic stability due to the addition of BS or other similar fillers, which implies that there is an increased interaction between the matrix and the reinforcement [19]. Nanofiller-modified composites have also shown improvements in thermal resistance and interfacial bonding but the results are not consistent with the conditions of processing and filler dispersion quality [20]. In most of these studies, however, properties are assessed almost individually. Mechanical performance, moisture resistance, and microstructural observations are usually measured and reported independently, and it is not easy to determine the interaction between these variables. The filler content, viscoelastic response, and interfacial morphological relationship in banana-sisal systems have not been adequately studied [21]. This lack of understanding is a practical issue. Despite the popularity of hybrid natural fiber composites as lightweight structures, their behavior over time under various mechanical and environmental conditions is not well characterized. In particular, the effects of BS on regulating the stiffness, damping, and moisture absorption of banana-sisal epoxy laminates have not been examined collectively. Another obstacle is obtaining homogeneous filler dispersion without agglomeration, which has a direct effect on load transfer and fracture behavior. In the absence of considering these factors, performance in laboratory conditions may not be successfully transferred to consistent performance in real-world applications [22]. This study aims to manufacture a banana-sisal hybrid epoxy composite with a controlled BS filler content and to examine the dependence of the filler concentration on the mechanical properties, viscoelastic behavior, and water absorption capabilities. This investigation is also aimed at correlating these responses with morphological characteristics to determine the contribution of interfacial bonding and filler distribution to governing composite performance.

MATERIALS AND METHODS

Banana and sisal fibers were used as reinforcement agents because they were light and considered good polymer composite supports. Agave sisalana provided banana fibers used on the pseudo-stem and sisal fibers, which were gathered on the leaves. Both were sourced as woven mats by a dealer in Coimbatore, India. The composition was designed to utilise the relatively flexible character of banana fibres and the increased rigidity of sisal fibres. Natural fibers, however, exhibit heterogeneity regarding quality and can be more sensitive to moisture, which can affect bonding and mechanical performance [23]. An epoxy system was selected as the matrix material to ensure that the fibers are well wetted and that they cure. The epoxy resin was bisphenol-A (LY 556) and a polyamine hardener (HY 951) at a ratio of 10:1, as stated by the supplier, Sakthi Fiber and Chemicals, Coimbatore. When working with plant fibers, epoxy is favored because of its bonding properties and dimensional stability; however, bonding depends on processing and fiber surface conditions. The filler added was BS powder to alter the stiffness and eliminate the effect of moisture on the matrix. The feed material was obtained through Otto Chemie Pvt. Ltd, Mumbai, whereby its average particle size was 44.5 µm. Filler was added at 0, 3, 6, and 9 wt%. The powder was dried at 80°C before being mixed to remove surface moisture. At higher loadings, even with drying, it is not easy to achieve uniform dispersion, and particle clustering can also affect the ultimate behavior of the composite [24]. Reinforcement was performed with banana (Musa spp.) and sisal (Agave sisalana) woven mats, and the laminates were made by a hand lay-up process. The surface of the mold was sponged and covered with polyvinyl alcohol such that the laminate could be peeled off without leaving any marks upon completion of curing. The order of stacking was predetermined, according to which the banana and sisal layers were alternated, keeping three layers of each to have a balanced structure. The epoxy resin was mechanically stirred with BS powder at approximately 700 rpm to attain decent dispersion. The mixture was allowed under a vacuum to remove trapped air. Once this was done, the hardener was poured in and stirred lower in a manner that would not create more bubbles. A thin coat of resin was initially applied to cover the mold, and the fiber mats were overlaid. Each layer was covered with resin that was sprayed with a brush and roller until it got beneath the fibers, and any air was forced out. Once lay-up was done, the laminate was lined with a release sheet and pressed with weights to ensure even pressing.

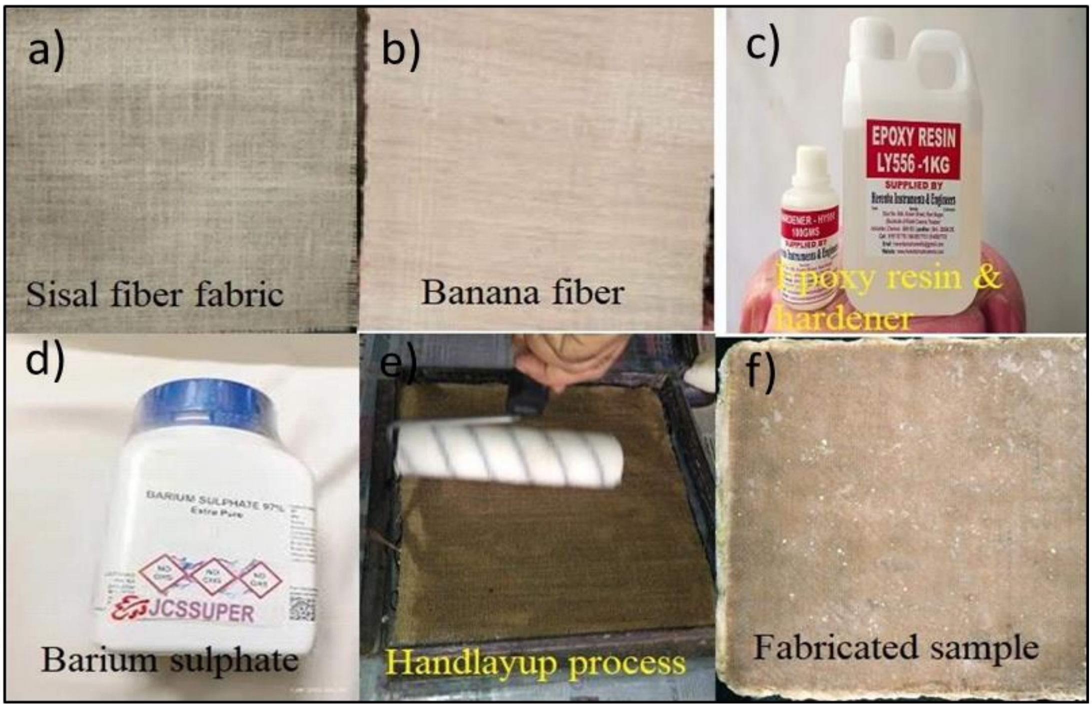

Figure 1. Materials and fabrication stages of banana/sisal BS reinforced epoxy composites showing (a) sisal woven mat, (b) banana fiber mat, (c) epoxy resin with hardener system, (d) barium sulfate microfiller, (e) laminate preparation through hand lay-up, and (f) cured composite laminate prepared for mechanical, dynamic, and water absorption characterization.

This enhanced the interaction between the fibers and matrix and minimized voids. The laminates were dried at room temperature for 24 h and then post-cured at 70°C for 3 h. The laminates were then cured and removed from the mold, trimmed at the edges, and cut into test specimens according to ASTM D638 and tested using a universal testing machine (FIE UTM-400, 400 kN capacity). A crosshead speed of 3 mm min-1 was used for the 250×25×3 mm specimens. A three-point bending test was conducted to determine the flexural properties with 127 mm × 12.7 mm × 3 mm samples having a span-to-depth ratio of 1:16. A Tinius Olsen IT504 pendulum impact tester (25 J capacity) was used to measure the Impact Strength (IS) according to ASTM D256 of specimens with similar dimensions [25]. The surfaces were hardened to a level of a 1.6 mm steel ball indenter according to ASTM D785 using a Mitutoyo HR-150A Rockwell hardness tester with a 1.6 mm steel ball indenter and a load of 100 kgf. In each composition, four specimens were tested, and the average values were considered [26]. A TA Instruments Q800 was used to perform a dynamic mechanical analysis in the dual-cantilever mode and a heating rate of 3°C in a temperature range of 30 to 150°C and a frequency of 1 Hz at different temperatures. The modes of storage, modes of loss, and damping behavior were recorded [27]. Scanning electron microscopy (JEOL JSM-IT300) was used to study the morphological characteristics of the fractured tensile samples. Before observation, the specimens were coated with gold to enhance conductivity and image clarity [28].

RESULT AND DISCUSSION

Tensile Test

The Tensile Strength (TS) of the hybrid laminates made of banana/sisal and BS fillers increased with increasing filler content up to 6 wt%, after which it slightly decreased with further increases in filler loading. The TS of the unfilled laminate was 72.8 MPa, which is a typical value for natural-fiber epoxy systems with moderate interfacial bonding and microvoids. The addition of 3 wt% BS increased the strength to 81.4 MPa. The TS reached a maximum of 89.7 MPa with 6 wt% filler, which is a definite improvement compared to the unfilled laminate. This tendency can be explained by the presence of BS particles in the matrix, which fill the open cavities and support the loading between the fibers and the resin [29]. It was also found that the synergistic interaction of banana and sisal fibers affected the response, as banana fibers can accommodate strain, but sisal fibers cannot deform under tensile loading. The TS decreased to 83.2 ± 0.12MPa at 9 wt% BS. This decrease can be attributed to a nonuniform filler distribution and clustering of particles, which create stress peaks and disastrous bonding around the periphery of the fiber. This observation is usually observed with natural-fiber composites at filler contents that are higher than the optimum. The laminate with 6 wt% BS exhibited the best tensile behavior of all the laminates, which implies that the levels of fillers and the arrangement of the fibers contribute to the overall tensile behavior. Figure 3. Figure 3 shows the change in TS of the banana/sisal-BS hybrid epoxy composite with the highest TS at 6 wt% filler, but with a minor decrease with increased filler content.

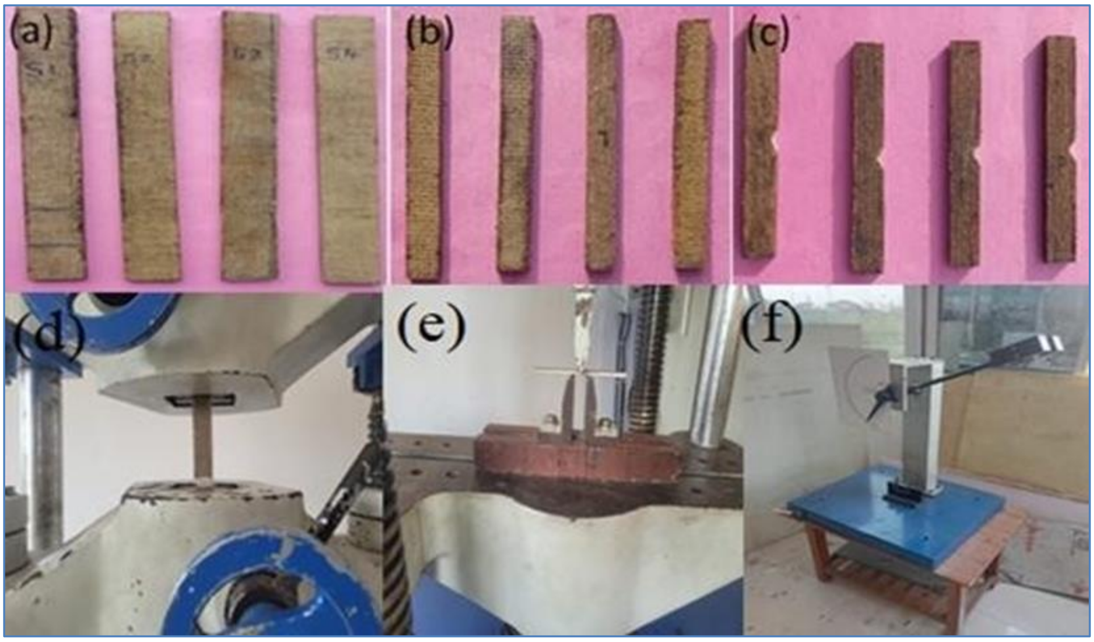

Figure 2. Experimental evaluation of banana/sisal BS hybrid epoxy composites showing specimens prepared for (a) tensile, (b) flexural, and (c) impact tests, along with the corresponding testing arrangements for (d) tensile loading, (e) three-point bending, and (f) pendulum impact testing carried out in accordance with ASTM standards.

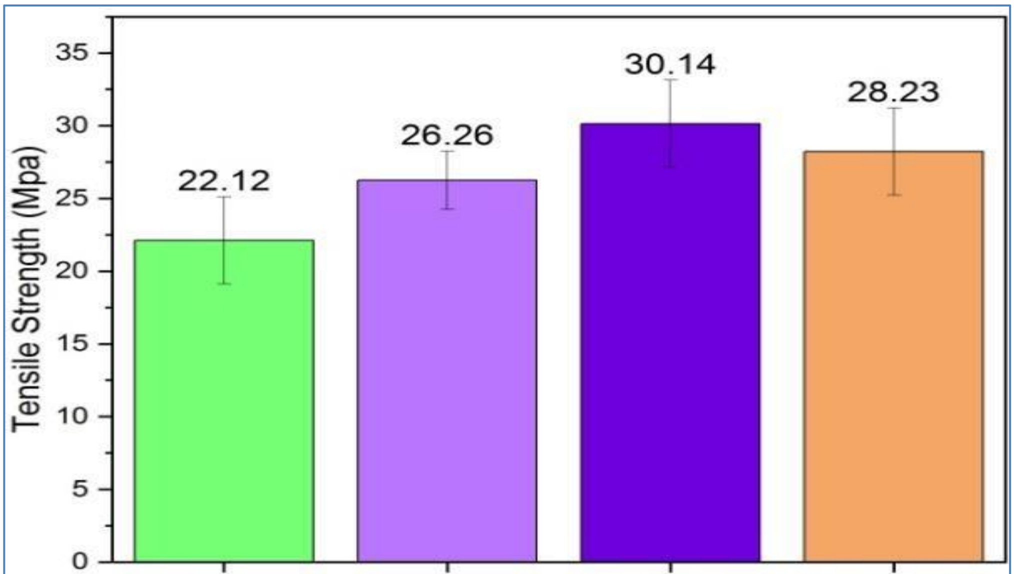

Figure 3. TS of hybrid composites.

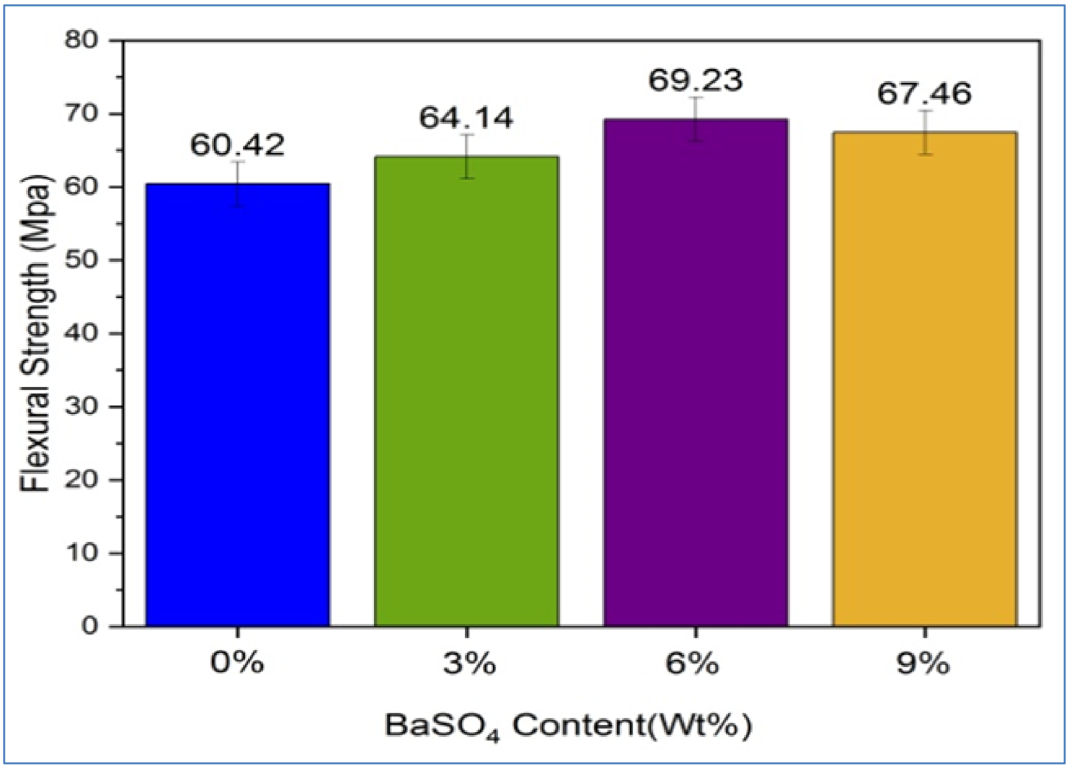

Figure 4. FS of hybrid composites.

FS

The FS increased with the addition of BS and exhibited a unique trend, increasing to 6 wt% and then slightly decreasing to 9 wt%. The FS of the unfilled laminate was 108.4 MPa. At 3 wt% BS, the value increased to 118.7 MPa, and the peak value of 128.9 MPa was achieved at 604.9 wt%. This enhancement can be explained by the fact that the filler is present in the epoxy and minimizes the areas of voids and helps to transfer loads between the layers of the fibers during bending. Further effects of the alternating arrangement of banana and sisal on the response were that the banana layers permitted limited deformation, but the sisal layers did not bend, and the laminate was able to support higher loads [30]. The strength decreased to 121.3 MPa with 9 wt% filler, which may be attributed to the poor distribution of particles and clustering in the matrix that formed weak areas under bending with the matrix. The same behavior is typically observed in natural-fiber laminates where the filler content exceeds the effective level. Across various compositions, the laminate with 6 wt% BS was more resistant to bending than the rest, indicating that the degree of filler and the structure of the fibers influence the flexural response. Figure 4 shows the FS of the banana/sisal-BS hybrid epoxy composite with its highest value at 60.13 wt% filler and a small decrease with increasing loading.

Impact Strength

The Impact Strength (IS) of the laminates made using banana/sisal/BS evidently increased with filler content up to 6 wt%, and then decreased slightly with filler content at 9 wt%. The unfilled laminate had an IS of 4.8 to 2 kJ/m^2. This was increased to 5.6 kJ/m^2 by the addition of 3 wt% BS and the highest strength of 6.3 kJ/m^2 was found at 6 wt.%. This enhancement can be explained by the effect of the BS particles in the matrix, which decrease the gap space in the matrix and hinder crack propagation during sudden loading. The fibrous structure was also helpful: banana fibers exhibited limited deformation and absorbed some of the impact energy, and sisal fibers did not fracture and maintained the integrity of the structure. Impeccability, at a filler concentration of 9 wt%, the IS was slightly reduced to 5.9 to 2 kJ, probably because of uneven particle distribution and possible clustering in the resin, which forms weak points in the impact. In all the compositions, the laminate with 6 wt% BS showed better impact performance; that is, both the filler percentage and the arrangement of the fibers have a synergistic effect on sudden loading resistance. The effect strength of banana/sisal with BS hybrid epoxy composites, as shown in Figure 5, shows that the maximum effect strength is at 6 wt% filler and there is a slight decline in the effect strength with an increase in the filler content.

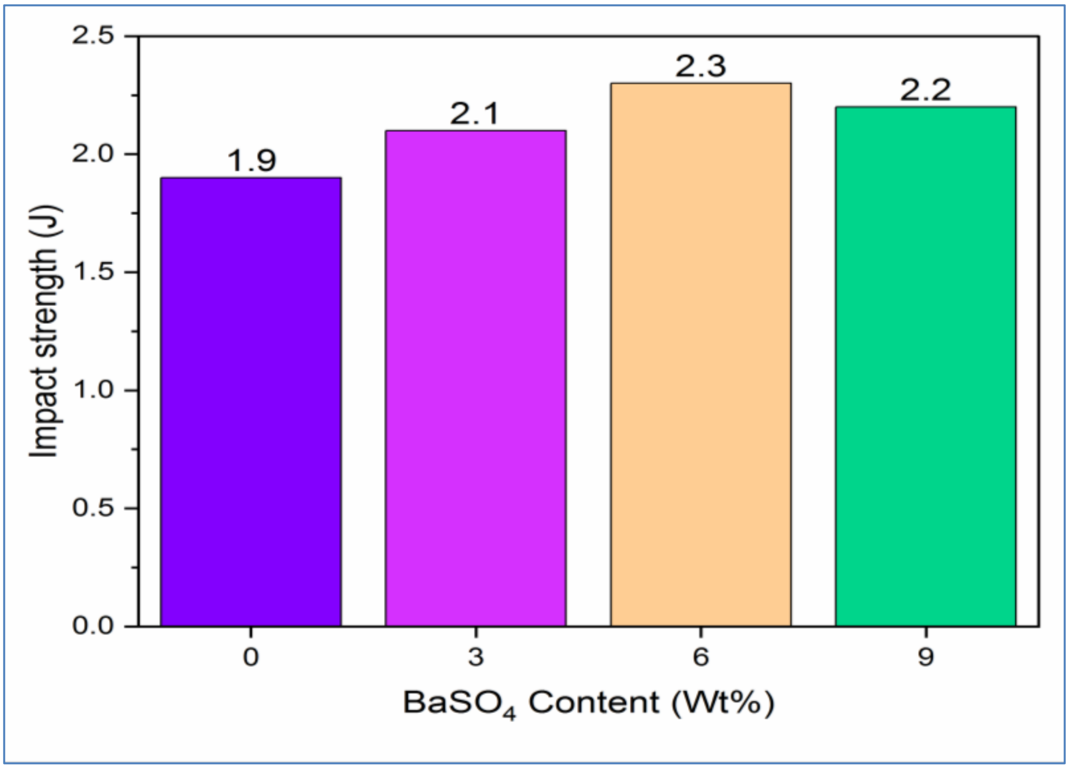

Figure 5. ISof the hybrid composite.

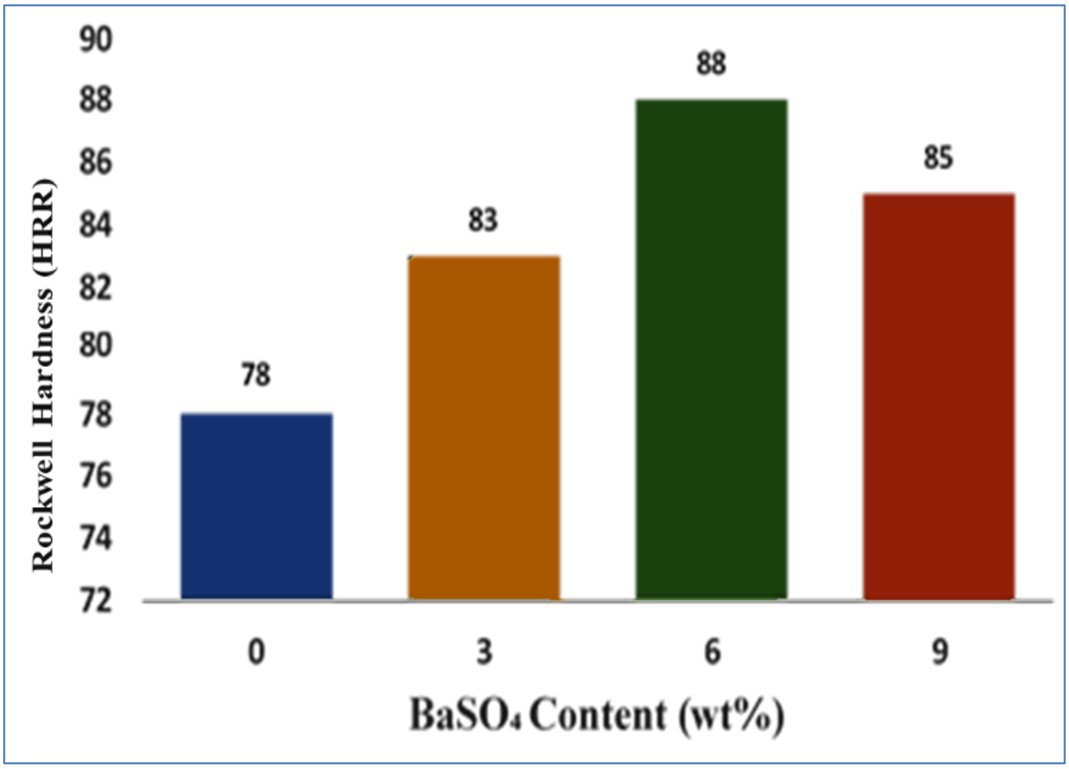

Figure 6. Hardness of the hybrid composite.

Hardness

The surface hardness increased with increasing BS content up to 6 wt%, and then marginally decreased at 9 wt%. The filler-free laminate had a hardness of 78 HRR. A BS content of 3 wt% increased the hardness to 83 HRR, with a peak at 6 wt%, reaching 88 HRR. This difference is mainly attributed to the fact that the epoxy matrix contains BS particles [31], which increases the indentation resistance and reduces the local deformation under load. The banana and sisal layouts also played a role in determining the results, as the load was spread throughout the laminate thickness. As the filler content rose to 9 wt%, the hardness dropped slightly to 85 HRR, which may be attributed to the agglomeration and rearrangements of particles within the metal, thus locally reducing the strength of the surface.

The sample that exhibited the best surface hardness among the assessed laminates was the 6 wt% BS, which is superior to the remaining laminates in terms of indentation resistance. Figure 6 shows the hardness of banana/sisal–BS hybrid epoxy composites at varying filler contents, where the hardness increased with increasing filler content and slightly decreased at higher filler contents.

Dynamic Mechanical Analysis

The viscoelastic behavior of banana/sisal hybrid laminates to which BS filler was added was studied using dynamic mechanical analysis. Differences in the storage modulus, loss modulus, damping factor, and glass transition temperature were measured to explain the stiffness and mobility of the molecules in the material. The unfilled laminate had a storage modulus of approximately 1.85 GPa. This parameter was enhanced when the mass of the solution was introduced with BS and reached 2.23 GPa at 6 wt%, thus displaying stiffer behavior. The storage modulus at 9 wt% decreased slightly to approximately 2.10 GPa, which can be due to an uneven distribution of the particles at the increased filler loading. The loss modulus also increased from 0.145 GPa to 0.171 GPa at 6 wt% and indicated an improvement in internal friction and energy dissipation. The damping factor showed a less pronounced tendency, decreasing to 0.070 at 6 wt% and 3-1 and increasing to 0.082 at 9 wt%, which could be explained by a more limited movement of the molecules; a slight increase was observed at 9 wt%, which can be potentially attributed to partial relaxation in the matrix. The laminate transition temperature changed to 94 ℃ at 6 wt% and then decreased slightly to approximately 90 ℃ at 9 wt%, indicating greater thermal stability at moderate filler concentrations. The structure of the fibers also affected the reaction: the presence of sisal layers led to rigidity, and the use of banana fibers allowed for minimal energy capture. The laminate with 6 wt% BS exhibited more stable behavior than that with other compositions. Figure 7 shows the storage modulus, loss modulus, tan, and glass transition temperature of banana/sisal–BS hybrid epoxy composites, highlighting the improved thermo-mechanical stability at 6 wt%.

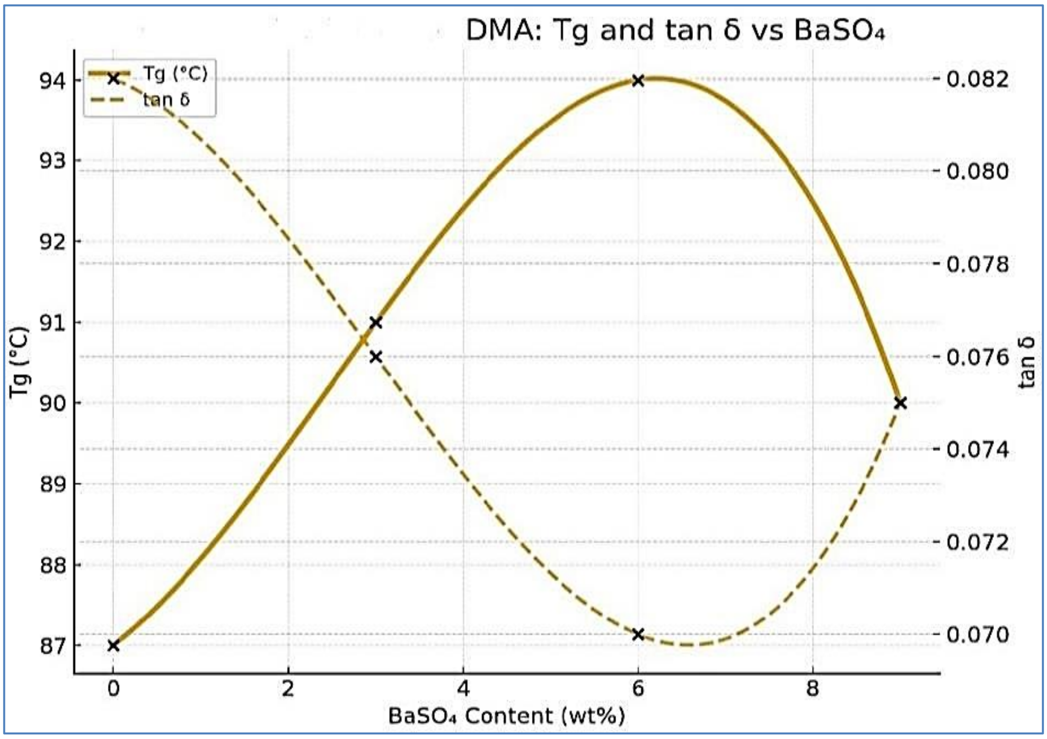

Figure 7. DMA Analysis of Hybrid Composites.

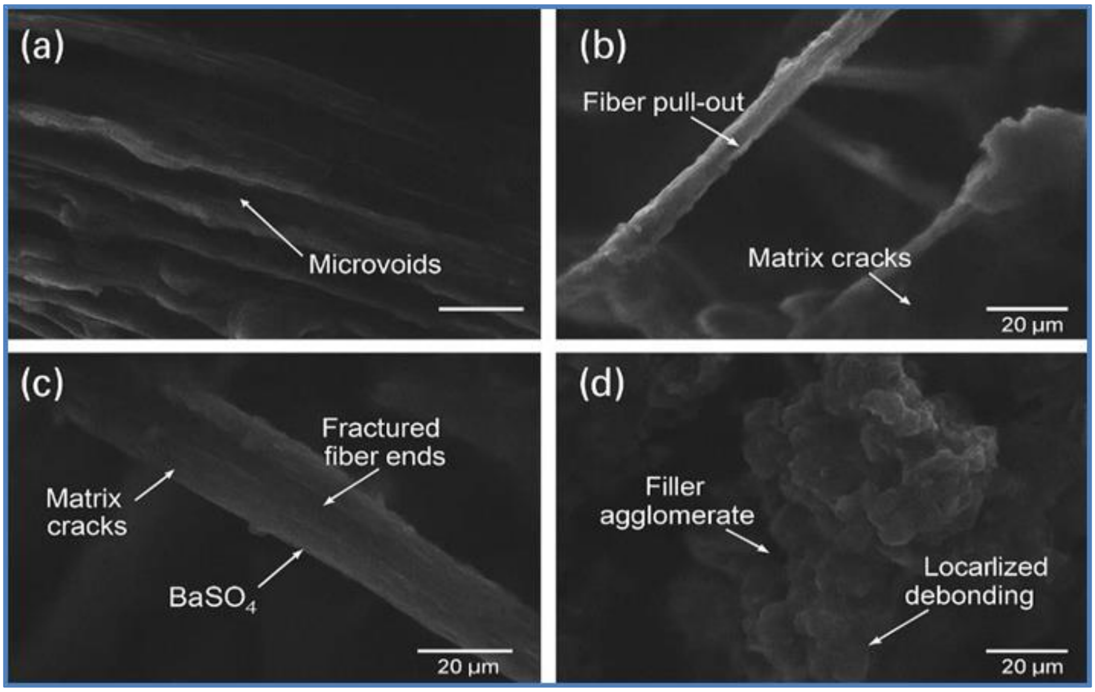

Figure 8. Microstructural Analysis of Hybrid Composites.

Microstructural Analysis

To clarify the effects of the addition of BS on the fiber–matrix interface and fracture morphology in banana/sisal epoxy laminates, scanning electron microscopy (SEM) of fractured tensile specimens was conducted. The uninfilled composite exhibited exaggerated fracture surface flaws, such as microvoids, matrix fissures, and conspicuous fiber pull-outs (Figure 8(a)). Fibers were evidently pulled out from the enveloping matrix at local points, implying that the wetting process was not optimally performed during fabrication. Therefore, there was inefficiency in the load transfer across the interface, which triggered an early tensile breakdown. Adding BS (0.03 g/kg) resulted in the creation of a smaller fracture surface. The strong bond between the resin and fibers was improved, and interfacial air pores were reduced (Figure 8(b)). Although fiber pull-out continued, the state of bonding was healthier than that in the unfilled laminate.

The sample containing 6 wt % BS exhibited a more compact morphology (Figure 8(c)). The dominant mode of failure was fiber breakage instead of pullout, and the interface was firmly bonded. The filler particles were visible along the fiber–matrix interface as well as in small voids, which suggested that they promoted stress transfer and reduced crack propagation [32]. This microscopic observation is in accordance with the improvements in the tensile and FS of this composition. However, SEM images can only be used to describe localized areas; therefore, it is pointless to make assumptions about the total uniformity over the entire laminate. When the BS content was 9 wt%, particle agglomeration and local cracks in the matrix were observed (Figure 8(d)). These features are likely to be stress concentrators and thus enhance crack initiation and interface separation. The nonuniform dispersion can therefore be blamed for the decline in mechanical performance at higher filler loadings. Similar trends have been documented in other natural fiber-based composites at filler concentrations above the optimum level. The combination of banana and sisal fibers also played a role in the overall effect: sisal was added to provide stiffness, and banana fibers were characterized by better deformation to failure, which could help to share the stress gradually. Nevertheless, the processing conditions and dispersion quality cannot be separated from these observations. Generally, the 6 wt% BS mix exhibited high interfacial integrity and lower defect density than the other laminates. There was less stability in the change beyond this level which can be attributed to uneven particle distribution. This was supported by the observations of the SEM. At a smaller filler concentration, fiber pull-outs and voids were apparent. The structure was found to be denser at 6 wt. with improved fiber embedding. Higher loading had agglomerated particles and microcracks. Such observations are made in the selected regions only and thus, they are tendencies and not the whole laminate. The response was also affected by the fiber combination. The stiffness was added by sisal and the banana fibres were more likely to deform with less fracture. This gave them a more evenly distributed load transfer in the laminate. This balance was further enhanced with addition of BS, especially at moderate level of filler content.

CONCLUSION

Banana/sisal-BS hybrid epoxy laminate was made through manual lay- up and compression molding, and their behaviour was investigated regarding mechanical behaviour, thermal stability, and interfacial properties. BS did not occur equally in any composition. The improvements were observed to a certain level, and beyond that, the response became random, which was primarily because of dispersion problems. There was a gradual increase of mechanical properties when fillers were added to 6 wt. The tensile strength was raised by 72.8 MPa to 89.7 MPa, and FS by 108.4 MPa to 128.9 MPa. The increase of the IS was also observed; it increased to 6.3 kJ/m^2. Hardness was also the same and attained 88 HRR. These evidences show that the filler helped in load sharing and limited local deformation in the matrix. The slight decrease in properties at 9 wt percent is probably due to clustering of the particles and creation of weak zones within the laminate and not due to any alteration in the fiber behaviour. Similar behaviour was observed in the thermomechanical response. The storage modulus was elevated and the glass transition temperature moved between 94 and 87 °C at the optimum filler content. This implies a decrease in the movement of the molecules within the matrix and an increased interaction between the fiber and the matrix. The 6 wt % composition exhibited the least change in behaviour in general, and has enhanced strength, stiffness and thermal resistance than the unfilled system. This renders it a potential aspirant of lightweight structural components and panels in which moderate weight carrying and impact-resistance is needed. The durability of the performance in the long term, moisture exposure and fatigue performance are to be thoroughly researched. The work in the future can involve treatment of fiber surface, finer fillers, and other resin systems in order to enhance consistency and service life.

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