Effect of fiber surface treatment and nanofiller on the Hardness behavior of Sisal fiber and Synthetic fiber reinforced with Polymer Matrix Hybrid Composites

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Abstract

This study investigates the fabrication and hardness performance of a hybrid epoxy composite reinforced with alkaline-treated sisal fibre, S-glass fibre, and graphene nanofiller. Sisal fibers were chemically treated using a 5% NaOH solution to enhance surface roughness and fibre–matrix adhesion, while S-glass fibers provided additional stiffness and load-bearing capability. Graphene nanoparticles were incorporated into the epoxy to improve surface hardness and reduce micro-cracking through enhanced interfacial bonding. The composite laminates were fabricated using the vacuum bagging technique, ensuring uniform consolidation and reduced void content. Shore D hardness testing was conducted in accordance with ASTM D2240. Results revealed that increasing S-glass content and graphene loading significantly improved hardness, with the highest value observed for the hybrid laminate containing 20% sisal, 20% S-glass, and 2% graphene. The combined effect of fibre hybridization, fibre treatment, and nanofiller reinforcement yielded a lightweight composite with superior surface resistance and structural stability. These improved hybrid composites show strong potential, making the optimized configuration highly suitable for applications that require superior surface strength, enhanced wear resistance, and reliable structural performance.

Keywords: *Sisal fibre, S-glass fibre, Graphene, Epoxy resin, Vacuum bagging, Shore D hardness.*

1. Introduction

Natural fibre reinforced polymer composites have gained significant attention due to their low cost, biodegradability, and favourable strength-to-weight ratio. Among natural fibres, sisal stands out for its high tensile strength, availability, and good adhesion characteristics when chemically treated. Alkaline treatment (NaOH) is widely used to remove lignin and hemicellulose, enhancing fibre roughness and improving fibre–matrix bonding [1]. However, natural fibres alone often lack the stiffness and surface hardness required for demanding engineering applications.

To overcome these limitations, hybridization with high-performance synthetic fibres such as S-glass has been introduced. S-glass fibre provides excellent tensile strength, superior stiffness, and improved thermal stability, making it an effective reinforcement in hybrid composites [2]. Epoxy resin acts as an ideal matrix due to its excellent adhesion, dimensional stability, and compatibility with both natural and synthetic fibres [3]. The addition of nanofillers such as graphene further enhances composite performance. Graphene exhibits exceptional mechanical strength, high surface area, and excellent load-transfer capability, enabling significant improvements in hardness and crack resistance [4]. Hybrid natural–synthetic fibre composites reinforced with nanofillers have shown significant improvements in hardness, stiffness, and overall mechanical behavior. Alkaline-treated fibers enhance bonding, while glass fibers increase rigidity. Incorporating graphene strengthens the matrix and improves crack resistance, making these hybrid composites suitable for durable, semi-structural applications.

2. Literature Summery

The development of natural fibre–reinforced polymer composites has received significant attention due to their sustainability, cost-effectiveness, and desirable mechanical properties. Among these, sisal fibre stands out for its high tensile strength, biodegradability, and availability.

Several studies have explored the impact of fibre treatment, hybridization, and filler incorporation on improving the hardness of sisal-based epoxy composites, which is a critical property influencing wear resistance, indentation behaviour, and surface durability.

Senthilkumar et al. (2024) investigated hybrid composites consisting of sisal and glass fibres reinforced in an epoxy matrix with SiC nanofillers. Their results showed that the combination of natural and synthetic fibres, along with nanofillers, significantly improved hardness, with 2 wt.% SiC producing the best performance. The improved hardness was attributed to effective nanofiller dispersion and strong fibre–matrix adhesion. This study is particularly relevant as it demonstrates the synergistic effect of hybrid fibres and nanofillers on surface properties [5].

Similarly, Sunil Kumar et al. (2025) reinforced sisal–epoxy composites with groundnut shell powder, reporting a marked increase in hardness with increasing filler content up to 40 wt.%. The rigid lignocellulosic powder restricted matrix deformation, resulting in improved surface resistance. This work highlights the role of agro-waste fillers in enhancing hardness, while maintaining low cost and sustainability. Fibre surface treatment has also been shown to influence hardness significantly [6].

Webo and Masu (2018) examined chemically treated and untreated sisal fibres and found that treated fibres produced higher hardness values. The enhanced interfacial bonding caused by alkali-silane-acid treatment improved fibre–matrix compatibility, reducing fibre pull-out and increasing resistance to indentation. This reinforces the importance of alkaline treatment for natural fibres [7].

Baseline studies, such as the one conducted by Haldar (2017), evaluated pure sisal–epoxy composites without fillers or hybrid fibres. Results showed a moderate improvement in hardness with higher fibre loading, though the enhancement was limited compared to hybrid or particle-filled composites. This suggests that sisal alone provides only incremental improvements in surface properties. Hybridization of natural fibres has also been explored [8].

Mosisa and Batu (2021) reported that bamboo–sisal hybrid composites exhibited increased hardness due to bamboo’s higher stiffness and hybrid synergy. This demonstrates that combining natural fibres can positively influence mechanical and surface characteristics [9]. In a similar context, Kumar et al. (2025) reinforced sisal–epoxy composites with groundnut shell particulates and found that hardness improved significantly, reaching up to ~104 HRL at 40 wt.% filler. This aligns with findings from other filler-based studies, confirming the effectiveness of particulate reinforcement [10].

Other studies support the effectiveness of hybrid natural–synthetic fibre composites and nanofillers in enhancing hardness and mechanical behavior. Joseph et al. (1999) reported that alkali-treated sisal fibres significantly improve fibre–matrix adhesion in thermoset composites [11]. Pothan et al. (2006) demonstrated that hybridizing sisal with glass fibre increases stiffness and reduces surface deformation [12]. Rafiee et al. (2009) found that graphene nanoplatelets considerably enhance hardness, modulus, and crack resistance in polymer matrices due to superior load transfer [13]. John and Thomas (2008) further highlighted that natural–synthetic hybrid composites exhibit improved durability, making them suitable for semi-structural applications [14]

Finally, Ram and Edwin Raj (2016) evaluated sisal–glass hybrid epoxy composites, reporting a substantial improvement in hardness due to the inherent stiffness of glass fibres. This study supports the notion that incorporating synthetic fibres can dramatically enhance surface durability [15]. Overall, the reviewed studies clearly show that fibre surface treatment, natural–synthetic fibre hybridization, and graphene-based nanofillers each play a critical role in enhancing composite performance. These findings justify the need for the present work aimed at developing a harder, stiffer, and more durable hybrid epoxy composite.

Author & Year	Composite System	Variables Studied	Hardness Findings	Relevance to Present Work
Senthilkumar et. 2024	Glass/Sisal fibres + epoxy + SiC nanfiller + SiC nanofiller	Nanofiller wt. (0–30%)	Maximum hardness at 2 wt. [illegible] n40 % nSiC	Closesly related: hybrid natural synethetic fibres
Sunil Kumar et. 2025	Sisal fitreated & untreated sisal fibres + epoxy	Filler content	Treated fibres produced higher hardness duto	Direct revenance to fibre treatment effects
Webo & Masu, 2018	Alkali-treated hybrid fibres + êpoxy	Surface treatment	Drected revenance to fibre treatment effects	Provees baseline sisal–epoxy hardness values
Mosisa & Batu, 2021	Bambou/Sisal hybrid fibres + epoxy	Hybrid ratios	Highest hardness with glass fibre	Supports hybridization effect on hardness
Ram & Edwin Raj, 2016	Sisal + Glass fibre hybrid epoxy	Filler wt. % at 40% filler	Highest hardness with glass fibre incorporation	Relevant to sisal–synthetic fibre hybrid behaviour

Figure 1. Some of the key findings from the literature review that support the present work

3. Materials Used

3.1. Natural fiber (Sisal Fiber)

Sisal fibre (Density: 1.3–1.5 g/cm³), highlighted across the reviewed studies, is a strong, biodegradable natural fibre widely used in polymer composites for its good tensile properties (400–700 MPa) and good impact resistance due to moderate toughness. The papers show that sisal improves hardness moderately on its own, but its performance significantly increases when combined with surface treatment, hybrid fibers, or fillers.

Chemical Properties

- Cellulose content: 65–78%
- Hemicellulose: 10–14%
- Lignin: 9–12%
- Rich in hydroxyl groups which improves reactivity with chemical treatments

3.2. Synthetic fiber (S-glass Fiber)

S-glass fibre (Density: 2.46 g/cm³) is a high-strength synthetic reinforcement known for its excellent tensile strength (4,500–5,000 MPa), stiffness, and thermal stability. It performs better than E-glass and offers superior resistance to heat, chemicals, and fatigue. When combined with natural fibers like sisal, it significantly enhances hardness, rigidity, and overall composite performance.

Chemical Properties

- Alumino-silicate composition with high silica content (Around 65%)
- Excellent chemical and corrosion resistance
- Stable in acidic and alkaline environments

3.3. Nano filler (Graphene)

Graphene (Density: 0.77 mg/m²) is a highly efficient nanofiller that significantly enhances composite hardness, stiffness, and interfacial bonding due to its exceptional strength and large surface area. When added to epoxy systems, graphene improves load transfer, reduces microcracks, and boosts mechanical performance, making it ideal for hybrid natural–synthetic fibre composites.

Chemical Properties

- Highly stable, chemically inert
- Strong π-π interaction with polymers
- Good dispersion after functionalization (e.g., oxidized graphene)

3.4. Polymer Matrix (Epoxy resin)

Epoxy (Density: 1.1–1.3 g/cm³) is a commonly used thermoset resin valued for its strong adhesion, high mechanical strength, and suitability with both natural and synthetic fibers. In composite applications, it increases hardness, enhances load transfer, and offers reliable thermal and chemical resistance. Its superior interfacial bonding makes it ideal for sisal–synthetic fibre–nanofiller hybrid composites.

Chemical Properties

- Highly crosslinked thermoset polymer
- Strong resistance to chemicals, solvents, and corrosion
- Excellent bonding with natural and synthetic fibres

4. Methodology

The hybrid composite laminate was fabricated using the vacuum bagging technique. Sisal fibers were first alkali-treated using a 5% NaOH solution for 4–6 hours, washed to neutral pH, and oven-dried at 60°C to improve surface roughness and fibre–matrix adhesion. S-glass fibers were cut to the required dimensions and arranged along with sisal fibers in a selected stacking sequence (alternative layer). Graphene-modified epoxy resin was prepared by dispersing graphene nanoparticles (1–2 wt.%) into epoxy using mechanical stirring, followed by addition of the hardener. The fibers were hand-laid and uniformly impregnated with the resin. Peel ply, breather fabric, and vacuum bag film were placed sequentially, and the assembly was sealed using tack tape. A vacuum pressure of –0.8 to –0.95 bar was applied to remove trapped air and ensure uniform consolidation. The laminate was cured under vacuum for 24 hours, followed by post-curing at 60–80°C to enhance the hardness and mechanical stability.

Figure 2. Pictorial of the fabrication process

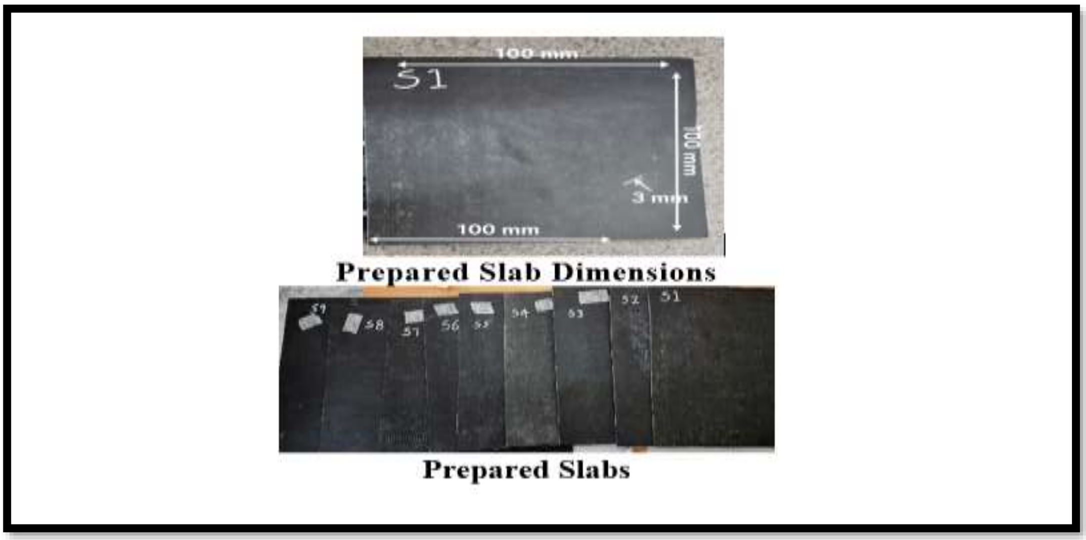

Figure 3. Composite slabs (Laminates) after fabrication process

4.1. Prepared sample specification

Composite laminates (9 different) measuring 100 mm × 100 mm × 3 mm were produced and machined with help of water jet machining to prepare the testing samples as per ASTM D2240 specifications for Hardness (Shore D) evaluation. The specifications of the fabricated composites are summarized in the table 1. below.

Table 1. Composite Slabs Specifications

Samples	Sample Combinations			
	Sisal fiber (wt.%)	S glass fiber (wt.%)	Epoxy resin (wt.%)	Graphene (wt.%)
1	40	0	59	1
	40	0	58.5	1.5
	40	0	58	2
2	30	10	59	1
	30	10	58.5	1.5
	30	10	58	2
3	20	20	59	1
	20	20	58.5	1.5
	20	20	58	2

4.2. Hardness test

The hardness of the fabricated hybrid composite specimens was evaluated using a Shore D durometer in accordance with ASTM D2240. The test measures the resistance of the material surface to indentation, providing an indication of its rigidity and surface strength. Since the standard requires a minimum thickness of 6 mm, the 3 mm composite sheets were stacked to achieve the required thickness while ensuring a smooth, flat testing surface. Each specimen was placed on a rigid base, and the indenter was pressed perpendicular to the surface with consistent force. Readings were taken after one second of indentation to ensure accuracy. A minimum of three measurements were recorded at different locations on each specimen, maintaining adequate spacing to avoid the influence of previous impressions. The final hardness value for each composite was obtained by averaging these readings. This test helps assess the effect of sisal treatment, S-glass reinforcement, and graphene addition on the surface behaviour of the hybrid composite.

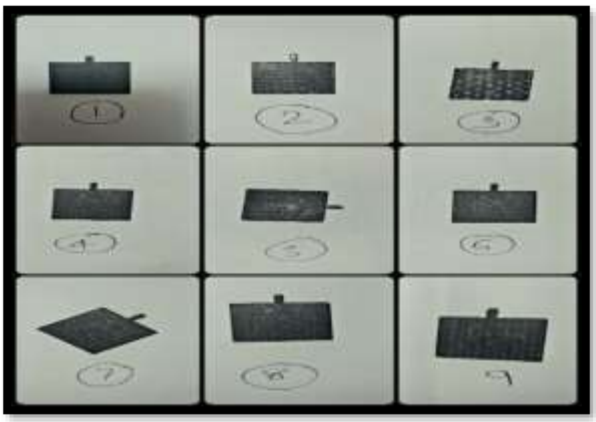

Figure 4. Samples for Shore D hardness testing

5. Results and discussion

Table 1. Results of the Shore D Hardness test

Samples	Sample Combinations				Hardness Number
	Sisal fiber (wt.%)	S glass fiber (wt.%)	Epoxy resin (wt.%)	Graphene (wt.%)	
1	40	0	59	1	50
	40	0	58.5	1.5	52
	40	0	58	2	53
2	30	10	59	1	65
	30	10	58.5	1.5	68
	30	10	58	2	70
3	20	20	59	1	82
	20	20	58.5	1.5	83
	20	20	58	2	84

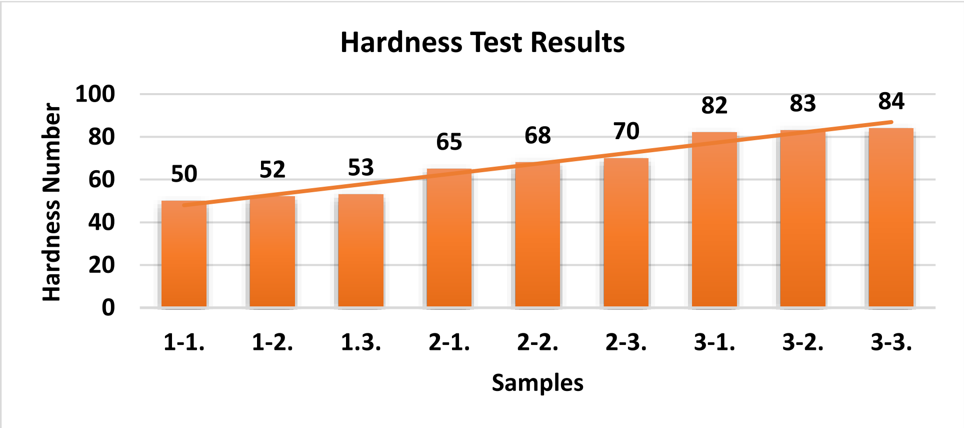

Figure 5. Bar graph Results of the Shore D Hardness test

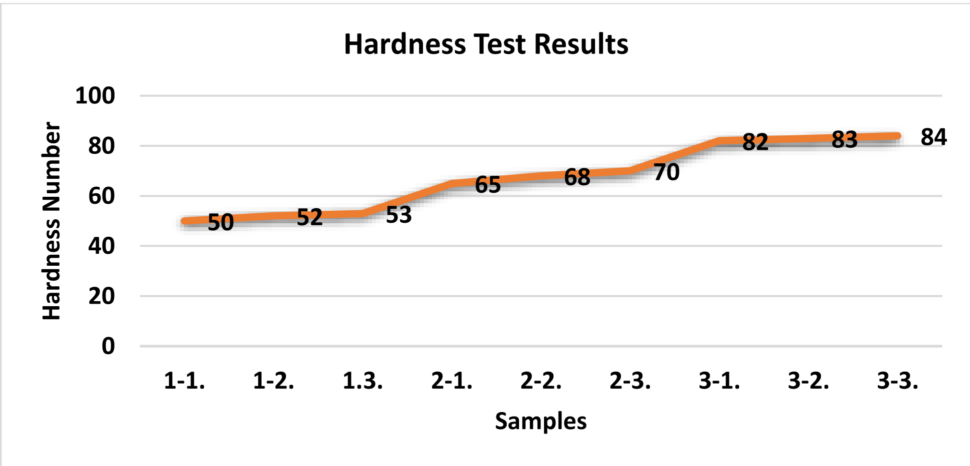

Figure 6. Line graph Results of the Shore D Hardness test

The above graphs (figure 5 and 6) represent the Shore D hardness values for hybrid composite samples prepared with varying proportions of alkaline-treated sisal fibre, S-glass fibre, epoxy resin, and graphene nanofiller. The results demonstrate a clear trend in which fibre hybridization and graphene loading significantly influence the surface hardness of the composites. In Sample Group 1, where the reinforcement consists only of sisal fibre (40 wt.%) and epoxy, hardness values range from 50 to 53 as graphene increases from 1 to 2 wt.%. This indicates that graphene improves surface rigidity, but the absence of synthetic fibres limits the overall hardness.

In Sample Group 2, replacing 10 wt.% sisal with S-glass results in a substantial increase in hardness values (65–70). This improvement is attributed to the higher stiffness, tensile strength, and load-bearing capability of S-glass fibres compared with sisal. The addition of graphene further enhances the indentation resistance through improved matrix–fibre bonding.

Sample Group 3 shows the highest hardness values (82–84) with an equal hybridization of 20 wt.% sisal and 20 wt.% S-glass. The synergy between the treated sisal fibres, stiff S-glass fibres, and uniformly dispersed graphene nanoparticles results in superior surface resistance. Overall, increasing S-glass content and graphene loading significantly enhances hardness, confirming their effectiveness in improving composite surface properties

6. Conclusion

The hardness assessment clearly shows that the combination of alkaline-treated sisal fibre, S-glass fibre, and graphene nanofiller significantly improves the Shore D hardness of the hybrid epoxy composites. Samples reinforced solely with sisal fibre exhibited moderate hardness values ranging from 50 to 53, indicating limited surface resistance. Introducing 10 wt.% S-glass resulted in a substantial improvement, with hardness increasing to 65–70, due to the superior stiffness and load-bearing capability of synthetic fibers. The highest hardness values, ranging from 82 to 84, were recorded for the composites containing an equal proportion of sisal and S-glass fibers (20% each) with increased graphene loading. This performance reflects the strong synergistic effect between the natural fibre, synthetic fibre, and graphene nanoparticles, which collectively enhance interfacial bonding and restrict micro-crack propagation. Overall, increasing S-glass content and graphene concentration leads to a much harder and more durable composite, making the optimized configuration highly suitable for applications requiring superior surface strength, wear resistance, and structural reliability.

7. Future Scope

The present study opens several opportunities for further research on hybrid natural–synthetic fibre composites. Future work can explore the influence of different chemical treatments or coupling agents on sisal fibers to further enhance interfacial bonding. The effect of varying graphene dispersion techniques, higher nanofiller loadings, or alternative nanoparticles such as nano-silica, alumina, or carbon nanotubes may also be investigated to optimize surface hardness and overall mechanical behaviour. Advanced fabrication techniques like resin infusion or compression moulding can be compared with vacuum bagging to study the influence of processing conditions on laminate quality. Additionally, finite element modelling (FEM) can be used to predict hardness and stress distribution within hybrid laminates. Exploring potential applications in automotive, marine, and structural components through prototype development and field testing will further validate the practical utility of these enhanced hybrid composites.

Acknowledgments

The authors sincerely thank Dr. H. K. Shivanand for his guidance and valuable suggestions throughout the work. Appreciation is extended to the SLN laboratory staff for their support during fabrication and testing, and to the institution for providing essential facilities and assistance that contributed significantly to the successful completion of this study.

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