

**MECHANICAL AND DYNAMIC-MECHANICAL
PERFORMANCE OF SUSTAINABLE COMPOSITES
REINFORCED WITH SISAL FIBERS**

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MASTER'S DISSERTATION IN MECHANICAL SCIENCES

**UNIVERSITY OF BRASÍLIA
FACULTY OF TECHNOLOGY
DEPARTMENT OF MECHANICAL ENGINEERING
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É justo que muito custe o que muito vale
Santa Tereza D'Ávilla

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ABSTRACT

MACEDO, M.S. Mechanical and Dynamic-Mechanical Performance of Sustainable Composites Reinforced with Sisal Fibers. Brasília, 2026. 88p. Master's Dissertation – Post graduate Program in Mechanical Sciences – Faculty of Technology, University of Brasília, Brasília, 2026.

Polymeric composites reinforced with natural fibers have emerged as promising alternatives to traditional synthetic materials, mainly due to their low density, renewable origin, and potential for reducing environmental impacts. However, the application of these materials in structural components still faces challenges related to the stability of their mechanical properties and their behavior under different loading conditions over time. In this context, it becomes essential to understand both the evolution of residual properties after long-term loading and the viscoelastic response under cyclic loading. Accordingly, this dissertation aims to investigate the residual mechanical and dynamic-mechanical behavior of sisal/epoxy composites and fiber-metal laminates (FMLs) hybrid-reinforced with sisal and synthetic fibers. In the first chapter, the residual thermal and mechanical properties of sisal/epoxy composites previously subjected to creep tests were evaluated, considering the influence of matrix curing conditions and fiber orientation. In the second chapter, the dynamic-mechanical behavior of sisal/epoxy composites with no prior loading history and of hybrid fiber-metal laminates was analyzed using dynamic mechanical analysis (DMA). The results showed a tendency toward residual matrix curing after creep, as reflected by an increase in the degree of cure and accompanied by improvements in thermal stability and, in certain cases, in the mechanical performance of the composites. For the systems with better interfacial quality, such as those subjected to room-temperature curing followed by post-curing, increases in tensile strength were observed after the test, on the order of 47.7% for the unidirectional laminates and 22.6% for the cross-ply laminates. Regarding dynamic-mechanical behavior, the viscoelastic response of the materials was strongly influenced by curing conditions, fiber orientation, and laminate architecture. The incorporation of natural fibers increased stiffness and altered dissipative mechanisms. In contrast, hybridization with synthetic fibers and the inclusion of metallic layers in the FMLs modified the stress distribution and the energy storage and dissipation capacity. It was also observed that the interaction between the different constituent phases plays a determining role in the dynamic response of the material, especially in hybrid systems. Overall, the results showed that the performance of sisal fiber-reinforced composites and hybrid fiber-metal laminates is directly related to the processing conditions, the quality of the fiber/matrix interface, and the structural configuration of the system.

Keywords: Dynamic mechanical analysis. Polymer composites. Sisal fiber. Sustainable materials. Residual properties.

RESUMO

MACEDO, M.S. Desempenho Mecânico e Dinâmico-Mecânico de Compósitos Sustentáveis Reforçados com Fibras de Sisal. Brasília, 2026 88p. Dissertação de Mestrado – Programa de Pós-Graduação em Ciências Mecânicas – Faculdade de Tecnologia, Universidade de Brasília, Brasília, 2026.

Os compósitos poliméricos reforçados com fibras naturais têm-se destacado como alternativas promissoras aos materiais sintéticos tradicionais, sobretudo em função de sua baixa densidade, de sua origem renovável e do potencial de redução dos impactos ambientais. No entanto, a aplicação desses materiais em componentes estruturais ainda enfrenta desafios relacionados à estabilidade de suas propriedades mecânicas e ao comportamento sob diferentes condições de carregamento ao longo do tempo. Nesse contexto, torna-se fundamental compreender tanto a evolução das propriedades residuais após solicitações de longa duração quanto a resposta viscoelástica sob carregamentos cíclicos. Diante disso, esta dissertação tem como objetivo investigar o comportamento mecânico residual e o comportamento dinâmico-mecânico de compósitos sisal/epóxi e de laminados de fibra-metal (FMLs) com reforço híbrido de fibras de sisal e de fibras sintéticas. No primeiro capítulo, foram avaliadas as propriedades térmicas e mecânicas residuais de compósitos sisal/epóxi previamente submetidos a ensaios de fluência, considerando a influência das condições de cura da matriz e da orientação das fibras. No segundo capítulo, foi analisado o comportamento dinâmico-mecânico de compósitos sisal/epóxi, sem histórico prévio de carregamento, e de laminados híbridos de fibra-metal, por meio de ensaios de análise dinâmica-mecânica (DMA). Os resultados mostraram uma tendência de evolução da cura residual da matriz após a fluência, refletida pelo aumento do grau de cura e acompanhada de melhorias na estabilidade térmica e, em determinados casos, no desempenho mecânico dos compósitos. Para os sistemas com melhor qualidade interfacial, como os submetidos à cura à temperatura ambiente, seguida de pós-cura, observaram-se aumentos na resistência à tração após o ensaio, da ordem de 47,7% para os laminados unidirecionais e de 22,6% para os laminados *cross-ply*. No que se refere ao comportamento dinâmico-mecânico, verificou-se que a resposta viscoelástica dos materiais é fortemente influenciada pelas condições de cura, pela orientação das fibras e pela arquitetura do laminado. A incorporação de fibras naturais promoveu um aumento da rigidez e alterações nos mecanismos dissipativos, enquanto a hibridização com fibras sintéticas e a inclusão de camadas metálicas nos FMLs modificaram a distribuição de tensões e a capacidade de armazenamento e dissipação de energia. Observou-se, ainda, que a interação entre as diferentes fases constituintes desempenha um papel determinante na resposta dinâmica do material, especialmente em sistemas híbridos. De forma geral, os resultados mostraram que o desempenho de compósitos reforçados com fibras de sisal e de laminados híbridos de fibra-metal está diretamente relacionado às condições de processamento, à qualidade da interface fibra/matriz e à configuração estrutural do sistema.

Palavras-chave: Análise dinâmico-mecânica. Compósitos poliméricos. Fibra de sisal. Materiais sustentáveis. Propriedades residuais.

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LIST OF ABBREVIATIONS AND SYMBOLS

A	– Adhesion factor
ASTM	– American Society for Testing and Materials
C	– Reinforcement efficiency
DMA	– Dynamic Mechanical Analysis
DSC	– Differential Scanning Calorimetry
E'	– Storage modulus
E''	– Loss modulus
E*	– Complex modulus
E' _g	– Storage modulus in the glassy region
E' _r	– Storage modulus in the flexible (rubbery) region
FML	– Fiber Metal Laminate
FRP	– Fiber Reinforced Polymer
NaOH	– Sodium hydroxide
PEEK	– Poly(ether ether ketone)
PLA	– Poly(lactic acid)
R	– Universal gas constant (8.314 J/mol·K)
Tan δ	– Damping factor
Tan δ_c	– Composite damping factor
Tan δ_m	– Matrix damping factor
T _g	– Glass transition temperature
TGA	– Thermogravimetric Analysis
T _m	– Melting temperature
V _f	– Fiber volume fraction
α_{cura}	– Degree of cure of the epoxy matrix
δ	– Phase angle
ΔE	– Activation energy
ΔH_{res}	– Residual enthalpy of the curing reaction
ΔH_{total}	– Total enthalpy of the uncured material
σ	– Stress
ε	– Strain

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1 INTRODUCTION

Over the past few decades, growing environmental concerns associated with the use of petrochemical-based materials have driven the search for more sustainable alternatives [1–3]. In this context, natural fiber-reinforced polymer composites have emerged as a promising option, particularly because they incorporate renewable resources as reinforcement within polymeric matrices [4,5]. Among these materials, lignocellulosic fibers, such as sisal, have gained considerable attention owing to their wide availability, biodegradability, and mechanical properties compatible with a variety of industrial applications [6,7].

Derived from the *Agave sisalana* plant, sisal fiber exhibits low density, high strength, and good energy absorption capacity. Its cellulose-rich composition contributes to increased material stiffness [8]. However, due to its hydrophilic nature, this fiber still exhibits limited adhesion to hydrophobic polymeric matrices, necessitating surface treatments, the incorporation of compatibilizers, or the optimization of processing conditions to improve fiber/matrix interfacial interaction [9].

Despite these advantages, the application of such materials in structural components still faces challenges related to mechanical durability and long-term property stability. Under real-service conditions, composites are frequently subjected to various types of loading, including long-term constant loads (creep) and cyclic stresses associated with variations in frequency and temperature [10]. In this regard, understanding the evolution of mechanical properties under such loading conditions is essential to ensure the reliability of these materials.

Creep plays a determining role in the evolution of the mechanical behavior of polymeric composites, being directly related to the viscoelastic nature of the matrix, fiber orientation, and the quality of the fiber/matrix interface [11]. Under prolonged loading, processes such as matrix degradation and interfacial deterioration may occur, compromising load transfer efficiency and, consequently, the structural performance of the material. On the other hand, depending on the loading conditions, creep may promote microstructural rearrangements and internal stress redistribution, resulting in changes in stiffness and system stability [12].

In this context, the evaluation of residual properties becomes essential for understanding the long-term behavior of composites. These properties are directly associated with the level of accumulated damage and the microstructural integrity of the material [13].

In natural fiber-reinforced composites, this analysis is particularly relevant, given that the hydrophilic nature of lignocellulosic fibers may compromise compatibility with polymeric matrices, reducing load transfer efficiency at the fiber/matrix interface.

In addition to long-term static loading conditions, the viscoelastic behavior of these materials can be investigated using dynamic mechanical analysis (DMA), which evaluates the material's response to cyclic loading as a function of temperature and frequency. Parameters such as the storage modulus, loss modulus, and damping factor provide relevant information on the system's stiffness, energy dissipation, and relaxation mechanisms [14]. Thus, DMA constitutes a fundamental tool for characterizing the viscoelastic behavior of composites, including those reinforced with natural fibers.

In this regard, several studies have contributed to advancing the understanding of the mechanical and dynamic-mechanical behavior of natural fiber-reinforced composites. Among them, the works of Teixeira *et al.* [15] and Libera Júnior [16] stand out as the foundation for the conception of this research. These studies highlight the importance of matrix-reinforcement interactions and the influence of processing conditions on the final properties of the materials.

Given this background, the present dissertation addresses two main topics, with sisal fibers as reinforcement as their common thread. In the first chapter, the residual thermal and mechanical properties of sisal/epoxy composites previously subjected to creep testing are evaluated. The analysis focuses on the influence of matrix curing conditions and fiber orientation on the residual behavior of the materials. The materials analyzed were originally developed and creep-tested by Teixeira *et al.* [15], with the present work focusing on investigating properties under the loading history.

In the second chapter, the analysis focuses on the dynamic-mechanical behavior of two distinct systems: sisal/epoxy composites subjected to different curing conditions and fiber orientations, without prior loading history, and hybrid fiber-metal laminates. The materials were provided by Teixeira *et al.* [15] and Libera Junior [16], respectively, and evaluated exclusively through DMA testing. The inclusion of hybrid laminates broadens the scope of the analysis by incorporating more complex systems, in which the interaction among different reinforcing phases directly influences the viscoelastic response of the material.

The integration of these two chapters enables a comprehensive analysis of the behavior of sisal fiber-reinforced composites, encompassing both the evolution of residual properties under long-term loading and the viscoelastic response under cyclic stresses. In

this way, the present work seeks to contribute to the understanding of the mechanisms governing the performance of these materials, with emphasis on the influence of curing conditions, laminate architecture, and fiber/matrix interfacial quality, fundamental aspects for the development of composites with stable structural performance and reliable long-term behavior.

1.1 OBJECTIVES

1.1.1 General Objective

To investigate the residual mechanical and dynamic-mechanical behavior of sustainable composites reinforced with sisal fibers and of fiber-metal laminates with hybrid reinforcement of sisal and synthetic fibers (carbon and glass).

1.1.2 Specific Objectives

- a) To investigate the residual thermal properties of sisal/epoxy composites and the epoxy resin, evaluating the effect of creep loading on the degree of cure of the matrix.
- b) To investigate the influence of matrix curing conditions and fiber orientation on the residual mechanical behavior of sisal/epoxy composites after creep testing.
- c) To investigate the dynamic-mechanical behavior of sisal/epoxy composites, considering the influence of matrix curing conditions and fiber orientation on the viscoelastic response of the material.
- d) To investigate the dynamic-mechanical behavior of fiber-metal laminates with hybrid reinforcement of sisal and synthetic fibers (carbon and glass), analyzing the effects of laminate architecture and aluminum incorporation on the viscoelastic response of the system.

CHAPTER I – EFFECT OF CREEP ON THE MECHANICAL AND THERMAL PROPERTIES OF SISAL FIBER-REINFORCED COMPOSITES

1.1 INTRODUCTION

The durability of natural fiber-reinforced polymer composites remains one of the primary challenges for their application in structural components. This is because conventional tests, which are generally short-term, do not always adequately represent the real loading conditions to which these materials are subjected throughout their service life. In practice, components are frequently exposed to different types of loading, including cyclic loads (fatigue), long-term constant loads (creep), and combinations thereof [17].

Among the different types of loading, creep stands out as particularly significant, characterized by the progressive deformation of the material under constant load over time [11]. In polymeric composites, this phenomenon becomes even more complex, as it results from the interaction between phases with distinct behaviors. The polymeric matrix, viscoelastic in nature, is more susceptible to deformation, while the fibers act as reinforcing elements, partially restraining it [18]. However, the efficiency of this mechanism depends strongly on the quality of the fiber/matrix interface and on microstructural parameters such as fiber orientation and the degree of matrix crosslinking. Thus, the composite response under creep cannot be understood solely from the individual properties of its constituents but rather from their interaction [19].

Beyond the accumulated deformation during loading, it is essential to evaluate how prolonged creep exposure affects the final state of the material. In this context, the analysis of residual properties becomes a fundamental aspect for understanding the durability and structural reliability of these composites. The evolution of these properties is directly related to damage mechanisms that develop within the material microstructure, such as fiber/matrix interfacial debonding, microcrack and microvoid formation, progressive degradation of the polymeric matrix, and possible alterations in fiber structure [20]. Additionally, phenomena such as fiber realignment and internal stress redistribution may occur during prolonged loading, processes that directly influence the mechanical stiffness and stability of the system [21].

Despite the relevance of the subject, significant gaps remain in the literature regarding the effects of creep on the residual properties of natural fiber-reinforced composites. A large portion of the available studies focuses on evaluating the behavior of these materials under hygrothermal aging or cyclic loading conditions, while investigations

directly correlating creep exposure with simultaneous changes in both mechanical and thermal properties remain relatively limited.

Given this scenario, this chapter aimed to investigate the effect of creep on the thermal and mechanical properties of sisal fiber-reinforced composites, considering different matrix curing conditions and fiber orientations. To this end, experimental analyses were conducted to evaluate residual properties using mechanical testing and thermal characterization techniques. Through this approach, the study sought to establish a direct correlation between creep-induced microstructural mechanisms and the evolution of material performance.

1.2 LITERATURE REVIEW

1.2.1 Natural Fiber-Reinforced Polymer Composites

The use of natural fibers as reinforcement in polymer composites has gained increasing prominence over the past few decades, driven primarily by environmental and economic demands [22]. Derived from renewable plant sources, natural fibers exhibit a lignocellulosic structure, composed primarily of cellulose, hemicellulose, and lignin, in addition to small fractions of pectins, waxes, extractives, and other secondary constituents [23].

From a structural standpoint, plant fibers can be understood as natural composites in which cellulose microfibrils are dispersed within an amorphous matrix composed of hemicellulose and lignin. Cellulose constitutes the primary component responsible for the stiffness and mechanical strength of the fiber, while hemicellulose and lignin contribute to structural cohesion, dimensional stability, and cell wall integrity [24].

Natural fibers can be classified by plant origin, with the most common obtained from stems, leaves, and fruits. This classification reflects variations in lignocellulosic composition and fiber morphology, factors that directly influence mechanical performance. Among the main plant fibers used as reinforcement in polymeric matrices, sisal, jute, flax, curauá, kenaf, and hemp are particularly noteworthy [25]. Table 1 presents the main physical and mechanical properties of these fibers.

Table 1 – Physical and mechanical properties of some of the main plant fibers used as reinforcement in polymer composites [22,24,26].

Fiber	Density (g/cm³)	Elongation (%)	Tensile Strength (MPa)	Elastic Modulus (GPa)	Cellulose Content (wt%)
Sisal	1.2–1.5	2.0–4.0	400–700	9–22	67–78
Jute	1.3–1.5	1.5–2.0	300–800	10–30	60–70
Curauá	1.4–1.5	2.0–4.0	600–1100	30–60	70–75
Hemp	1.4–1.5	1.6–3.0	550–900	30–70	60–75
Kenaf	1.2–1.5	1.5–3.0	220–930	14–53	45–57
Flax	1.4–1.5	1.2–3.0	500–1500	30–60	30–70
Pineapple	1.4–1.5	1.0–3.0	400–1600	34–82	70–82
Coconut	1.1–1.2	15–25	100–250	2.0–6.0	32–43

Natural fibers offer several advantages when used as reinforcing elements in polymeric matrices, from both technical and socioenvironmental standpoints. One of the

primary advantages is their low density, ranging from 1.2 to 1.5 g/cm³, significantly lower than that of synthetic fibers, enabling the production of lighter materials with good mechanical properties [27].

Furthermore, because they are derived from renewable sources, natural fibers help reduce environmental footprints and support the development of more sustainable technologies [26]. Another relevant aspect is the low cost of these fibers, resulting from their wide availability and the possibility of utilizing agro-industrial waste, which also supports the strengthening of local and regional production chains [28].

In terms of mechanical performance, some fibers, such as curauá, pineapple, flax, and hemp, exhibit high elastic modulus, enabling the development of composites with competitive stiffness compared to those reinforced with synthetic fibers. This characteristic is primarily associated with the high cellulose content and the high degree of microfibril orientation along the fiber axis, both of which directly influence the elastic response of the material [29].

Despite these advantages, natural fibers present technical limitations that must be considered. The primary restriction is their high hygroscopicity, arising from hydroxyl groups in the cellulose chains that promote hydrogen bonding with water molecules and, consequently, moisture absorption. This phenomenon compromises the dimensional stability of the material, as water absorption promotes fiber swelling and alters the characteristics of the fiber/matrix interface. Moreover, the presence of moisture at the fiber/matrix interface tends to reduce interfacial adhesion efficiency, impairing load transfer and, consequently, the mechanical performance of the composite [30,31].

Another limiting factor is the intrinsic variability in the physical and mechanical properties of these fibers, which is influenced by cultivation conditions, plant age, extraction methods, and geographic location. This variability leads to high structural heterogeneity, which hinders standardization in industrial applications and compromises the predictability and reliability of composite mechanical performance [32]. In addition, natural fibers exhibit relatively low thermal resistance, with the onset of degradation around 250 °C, due to the decomposition of their lignocellulosic constituents, particularly hemicellulose and cellulose, which limits their use in thermoplastic matrices processed at high temperatures and in systems requiring greater thermal stability [33,34].

To overcome these limitations, various strategies have been developed to improve the stability, compatibility, and durability of these materials. One of the most effective approaches involves applying chemical treatments to fiber surfaces. Processes such as

alkalization with sodium hydroxide (NaOH), acetylation, and benzylation are used to remove impurities, reduce hygroscopicity, and enhance adhesion to the polymeric matrix [35]. The use of coupling agents, such as functional silanes, has also proven effective, promoting chemical interactions between the reinforcement and the matrix and increasing the interfacial strength of the composite [36].

Another relevant strategy is hybridization, which combines natural and synthetic fibers within a single polymeric matrix. This technique allows exploitation of synergistic effects among different types of reinforcement, with synthetic fibers contributing to mechanical strength and stiffness. In contrast, natural fibers reduce the cost and density of the material [37]. Furthermore, hybridization can mitigate typical limitations of plant fibers, such as variability in properties and lower thermal stability, thereby contributing to greater predictability of mechanical performance and higher structural reliability of the composite, without eliminating the socioenvironmental benefits associated with natural fibers [38].

In this context, natural fiber-reinforced polymer composites have been gaining prominence across various industrial sectors, driven by environmental requirements, the need to reduce component weight, and technological advances in sustainable materials processing. The combination of versatility, mechanical strength, and lightweight characteristics has enabled the incorporation of these materials into different industrial segments [39].

In the aeronautical sector, although still restricted to non-structural components, composite materials have been employed in cabin interior linings, luggage compartments, and finishing elements. Their low density makes these materials particularly suitable for applications where weight reduction is a determining factor for system energy efficiency [40]. Although the use of primary structural elements still requires greater control of variability and long-term performance evaluation, studies indicate that hybridization of natural and synthetic fibers can achieve mechanical performance adequate for structural components [41].

In the automotive industry, these materials are widely used in the manufacture of non-structural interior components, such as door panels, roof liners, seat backrests, and instrument panel finishing elements [42]. For more than two decades, automakers such as BMW, Mercedes-Benz, Audi, Ford, Volkswagen, and General Motors have been incorporating natural fiber-reinforced composites as full or partial replacements for fiberglass composites, aiming at weight reduction, increased energy efficiency, and lower carbon dioxide emissions throughout the vehicle life cycle [43].

In the construction industry, natural fiber-reinforced composites are used to manufacture partition panels, cladding panels, decorative elements, extruded profiles, roof tiles, sandwich panels, and sustainable furniture [44]. In these systems, thermoacoustic performance plays a central role owing to the low thermal conductivity of the lignocellulosic phase and the high capacity for acoustic energy dissipation [45]. Furthermore, the incorporation of plant fibers as reinforcement in polymeric or cementitious matrices enables the utilization of agroindustrial waste, contributing to the reduction of environmental impacts and to the development of more sustainable construction practices, particularly in regions with lower levels of industrialization [46].

In the packaging sector, composites based on natural fibers and biodegradable matrices have been investigated as alternatives to conventional petrochemical-based materials, particularly in short-service-life applications. The incorporation of lignocellulosic reinforcement into biodegradable polymers enables the development of materials with lower environmental persistence, while maintaining mechanical performance compatible with the functional requirements of packaging and protection [47].

Similarly, these systems are also used in the manufacture of sporting goods and durable consumer goods, such as bicycle frames, skateboards, helmets, snowboards, musical instruments, and toys. In these cases, low density, combined with stiffness and mechanical strength, enables the production of lightweight products with adequate structural performance [48].

In the biomedical field, recent studies have investigated the use of natural fibers and their composites in the development of medical textiles, sutures, orthopedic implants, and tissue-engineering scaffolds. The biocompatibility, biodegradability, and antimicrobial properties of these fibers have been identified as promising characteristics for clinical applications, particularly in preventing infections associated with medical devices [49]. Furthermore, incorporating these fibers into polymeric matrices enables the production of biocomposites with good mechanical properties and potential applications in orthopedics and tissue regeneration, although *in vivo* studies are still required to validate their safety and clinical performance [50] fully.

Thus, natural fiber-reinforced polymer composites are consolidating their position as a promising alternative for a wide range of applications. Despite the inherent challenges associated with the lignocellulosic nature of the fibers, advances in surface modification techniques and interface engineering have improved the performance and reliability of these systems. This progress is expected to gradually broaden their field of application,

strengthening their role as a strategic material in developing solutions aligned with the demands of sustainability and technological competitiveness.

1.2.2 Effect of Creep on the Residual Properties of Natural Fiber-Reinforced Polymer Composites

The creep test is a widely used technique for evaluating the mechanical response of materials under constant load over time. Unlike conventional mechanical strength tests, in which the load is applied incrementally until failure, the creep test allows for the analysis of the evolution of material deformation under constant loading [51,52]. In polymeric composites, this test is particularly relevant, as it enables investigation of the combined behavior of the matrix and the reinforcement, as well as the assessment of the mechanical performance of the material over time.

In general, the creep test presents three main stages. In the primary creep stage, the strain rate is high but decreases gradually as the material structurally adjusts to the applied stress. Subsequently, the material enters the secondary creep stage, in which the strain rate tends to stabilize and remain approximately constant over time. Finally, in the tertiary creep stage, a rapid increase in strain is observed, followed by material failure [53,54]. Figure 1 schematically illustrates the typical creep behavior of viscoelastic materials over time, highlighting the three stages described.

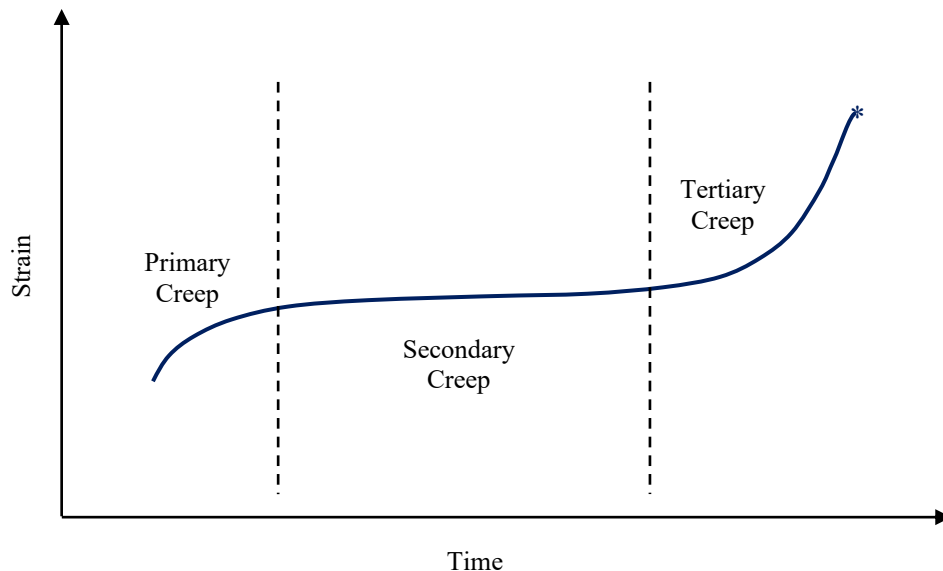


Figure 1 – Typical creep strain behavior over time. Adapted from Liu *et al.* [53] and Katouzian *et al.* [54].

When a polymeric material is subjected to mechanical stress, part of the deformation occurs immediately and is associated with the elastic response of the molecular network.

However, due to the mobility of the polymer chains, molecular rearrangement processes also occur more slowly. These rearrangements occur progressively over time, allowing for the gradual reorganization of chain segments into energetically more stable configurations. Thus, the deformation of the material is not limited to the instant of load application, but continues to evolve progressively, characterizing a time-dependent behavior typical of viscoelastic materials [55].

When these polymeric materials are used as a matrix in natural fiber-reinforced composites, the creep behavior becomes more complex due to the presence of two phases with distinct mechanical properties. The polymeric matrix, exhibiting a more pronounced viscoelastic behavior, tends to undergo progressive deformation under constant stress over time. Natural fibers may also exhibit viscoelastic response; however, due to their cellulose-rich structure, the time-dependent deformation is significantly lower. Thus, when the composite is subjected to prolonged loading, the matrix tends to concentrate most of the deformation. At the same time, the fibers act as reinforcing elements, restraining this deformation and contributing to the mechanical stability of the material [56].

The deformation restraint provided by the fibers is related to the load-transfer mechanism from the matrix to the reinforcement, which helps reduce creep rate of the material. However, the efficiency of this process depends directly on the quality of the fiber/matrix interface. An interface with good adhesion favors stress transfer from the matrix to the fibers and contributes to improved mechanical performance of the composite. On the other hand, when interfacial adhesion is limited, phenomena such as fiber/matrix debonding, interfacial sliding, and microvoid formation may occur, reducing load transfer efficiency and potentially intensifying creep deformation, thereby compromising the long-term mechanical performance of the material [57].

Furthermore, the effect of creep on the mechanical performance of the material depends on several factors, including the type of matrix, reinforcement content, fiber orientation, and environmental conditions (temperature and humidity) [58]. When fibers are well aligned in the loading direction, and an efficient interface exists between the matrix and the reinforcement, greater restraint of the viscoelastic deformation of the matrix is achieved, resulting in lower creep rates. In contrast, random arrangements or angular fiber orientations tend to exhibit greater deformation under constant load, due to the lower efficiency in restraining matrix movement [59].

The curing conditions applied to the matrix also play a significant role in the creep behavior of composite materials. The curing process is directly related to the formation of

the three-dimensional network of the polymeric matrix, which results from crosslinking reactions between chains. Matrices with a higher degree of cure tend to exhibit denser polymer networks, which limit molecular mobility and reduce susceptibility to deformation. In contrast, systems with lower crosslink density may exhibit greater polymer chain mobility, which contributes to deformation under long-term constant loading [60].

In this regard, it is important to evaluate not only the deformation developed during the creep test but also the mechanical state of the composite after the period of exposure to a constant load. Thus, residual properties correspond to the mechanical properties that remain in the material after it has been subjected to real service conditions [12]. In polymers and polymeric composites, these properties are directly related to the level of accumulated damage in the material microstructure, the magnitude of which may cause changes in the mechanical behavior of the composite, with or without a reduction in its mechanical properties.

In general, the residual behavior of composite materials depends on several factors, including loading conditions (stress level and exposure time), environmental conditions (temperature and humidity), and the material architecture, such as fiber orientation and fiber/matrix interfacial quality [61]. Interface integrity plays a particularly important role in this process, since efficient load transfer between the matrix and the reinforcement helps limit the viscoelastic deformation of the matrix and reduce damage accumulation in the material's microstructure. Thus, composites with good interfacial adhesion and properly oriented fibers tend to exhibit lower creep deformation and, consequently, better retention of mechanical properties after prolonged loading [62].

Several studies report that critical service conditions can significantly degrade the residual properties of polymeric composites. In general, increased loading exposure time and more severe environmental conditions intensify the damage mechanisms in the material. Under such conditions, processes such as plasticization of the polymeric matrix, degradation of natural fibers, and weakening of the fiber/matrix interface may occur, resulting in reductions in mechanical strength and elastic modulus after the exposure period [63].

Experimental results demonstrating this relationship were reported by Feng *et al.* [61], who investigated the creep behavior and residual mechanical properties of glass fiber-reinforced polymeric composites. The authors observed that after 1000 hours of loading at 70% of the ultimate stress, there was a slight reduction in residual properties, with decreases of approximately 6.6% in tensile strength and 6.2% in elastic modulus. These results were attributed to the progressive accumulation of microstructural damage, including fiber

breakage, microcrack formation in the polymeric matrix, and degradation of the fiber/matrix interface. Furthermore, the authors observed an increase in material porosity after the creep test, which contributed to the reduction in residual mechanical properties.

On the other hand, the literature indicates that under certain loading conditions, residual mechanical properties may exhibit negligible variation or even slight increases. This behavior has been associated with possible fiber realignment during load exposure, as well as improved stress transfer and distribution within the material. Furthermore, microstructural rearrangements may, in certain situations, contribute to increases in residual stiffness or strength, particularly when the integrity of the fiber/matrix interface and the reinforcing fibers is preserved [13].

Studies by Ai *et al.* [12] confirmed this trend by evaluating the residual mechanical properties of carbon fiber-reinforced polymeric composites after 1000 hours of creep exposure. The results demonstrated that at lower stress levels, residual strength was not only preserved but also increased slightly relative to the initial values, a behavior associated with progressive fiber alignment during loading. In contrast, at higher stress levels, a reduction in residual strength was observed, attributed to the formation of microstructural defects, such as discontinuities at the fiber/matrix interface and possible fiber damage. Furthermore, the residual elastic modulus remained essentially unchanged across all evaluated conditions, indicating that the material's stiffness was not significantly affected by prolonged loading.

Overall, studies in the literature indicate that creep can significantly influence the residual properties of polymeric composites, with this response directly associated with the damage mechanisms that develop in the material's microstructure during loading. It is observed that, depending on stress conditions, exposure time, and the integrity of the fiber/matrix interface, residual properties may range from moderate reductions to relatively minor variations.

However, most investigations have focused on synthetic fiber-reinforced composites, and the understanding of the residual behavior of natural fiber-reinforced composites remains limited. Moreover, the influence of factors such as matrix curing conditions and fiber orientation in this context has not yet been fully established, which reinforces the need for further studies correlating viscoelastic behavior with the evolution of residual properties of these materials.

1.3 MATERIALS AND METHODS

1.3.1 Materials

Composite materials used in this study were produced and creep-tested in a previous work developed by Teixeira *et al.* [15]. Overall, the composites were manufactured by compression molding using Araldite® LY 5052 epoxy resin and Aradur® 5052 hardener at a 100:38 ratio, with a 50% volume fraction of sisal fibers. The fibers were previously treated in a 5% (w/v) aqueous NaOH solution, at a solution-to-fiber ratio of 10:1, at 50 °C for 2 h under manual stirring, and subsequently oven-dried.

Two groups of materials were manufactured: unidirectional composites with a [0°] orientation and cross-ply laminates with a [0°/90°/90°/0°] orientation. The samples were subjected to three distinct curing routes: (i) curing at room temperature for 24 h, followed by post-curing in an oven at 120 °C for 1 h; (ii) curing in an oven at 100 °C for 4 h; and (iii) curing at room temperature with the addition of an accelerator at a 100:38:1 ratio, without subsequent post-curing. For comparison, unreinforced epoxy matrix samples were also produced and subjected to the same curing conditions as the composites. Table 2 presents the main characteristics of the analyzed materials.

Table 2 – Characteristics of the materials subjected to the residual tensile test, including the epoxy matrix and sisal/epoxy composites, under different curing conditions and fiber orientations. Adapted from Teixeira *et al.* [15].

Material	Curing Condition	Dimensions (mm)			Creep Stress (MPa)
		Length	Width	Thickness	
Epoxy Matrix	Room temperature cure with post-cure	193.0	19.1	2.5	14
	Oven cure at 100 °C/4 h	196.0	19.6	2.3	
	Cure with accelerator	197.0	20.9	2.5	
Sisal/Epoxy [0°]	Room temperature cure with post-cure	194.0	23.5	3.1	32
	Oven cure at 100 °C/4 h	184.0	22.4	2.7	
	Cure with accelerator	189.5	21.8	2.6	
Sisal/Epoxy [cross-ply]	Room temperature cure with post-cure	201.0	22.9	3.0	16
	Oven cure at 100 °C/4 h	179.0	22.7	3.1	
	Cure with accelerator	190.0	22.4	3.8	

The tensile creep test was performed in the laboratory under controlled conditions at 24 ± 2 °C and a relative humidity of 45–55% for 24 h. The creep stresses applied to the materials were 14 MPa for the epoxy matrix, 32 MPa for the sisal/epoxy [0°] composite, and 16 MPa for the sisal/epoxy [cross-ply] composite, corresponding to approximately 40% of the ultimate tensile strength of each material group. Due to the long duration of each test and the limited availability of equipment, only one sample was tested per experimental condition.

1.3.2 Mechanical Characterization by Residual Tensile Testing

The residual tensile tests were conducted in accordance with the guidelines established by ASTM D3039 [64]. The tests were performed on an Instron 8801 universal testing machine equipped with a 100 kN load cell, available at the Mechanical Testing Laboratory of the Faculty of Engineering Sciences and Technologies – Campus UnB Gama. The specimens were gripped with wedge grips adjusted to a contact pressure of 1 MPa, and the test speed was set to 1 mm/min. An Instron extensometer was also employed to determine specimen strain. Figure 2 presents the experimental setup used to conduct the tests.

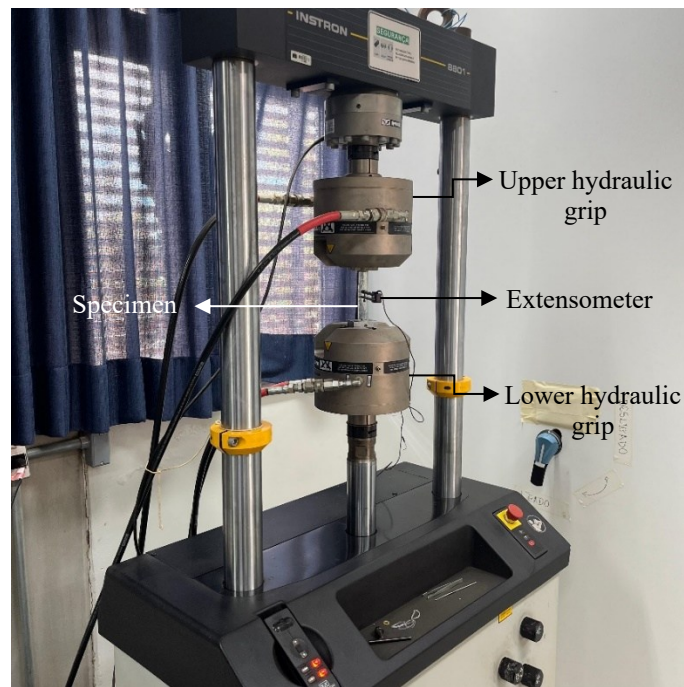


Figure 2 – Experimental setup of the residual tensile test.

The residual tensile test was performed to evaluate the mechanical behavior of the material after prolonged exposure to constant loading. For this purpose, mechanical characterization was conducted on specimens previously subjected to creep, which already presented a prior history of mechanical loading. It should be noted that creep tests require long execution periods, which, combined with the limited availability of specimens during

the development of this research, made experimental replicates unfeasible. Accordingly, only one specimen was tested for each experimental group.

1.3.3 Residual Thermal Characterization by Thermogravimetry (TGA) and Differential Scanning Calorimetry (DSC)

Following the residual tensile testing, the specimens were thermally characterized using thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC). TGA tests were performed in accordance with the guidelines of ASTM E2550 [65]. The analyses were performed on samples of epoxy matrix and of the epoxy composite reinforced with unidirectionally oriented sisal fibers at 0°. Since thermal analysis techniques require small amounts of material, fiber orientation does not significantly influence the results. Therefore, sisal/epoxy composite samples with a cross-ply configuration were not evaluated.

The tests were performed using a TA Instruments SDT Q600 simultaneous thermal analyzer, available at the Instrumental Analysis Laboratory of the Faculty of Engineering Sciences and Technologies – Campus UnB Gama. The tests were conducted in duplicate, using approximately 10 mg of sample placed in alumina crucibles and subjected to controlled heating.

The samples were heated from room temperature to 600 °C at a heating rate of 10 °C/min, under a nitrogen atmosphere with a gas flow rate of 100 mL/min. Based on the results obtained, it was possible to determine the degree of cure of the epoxy matrix and of the sisal/epoxy [0°] composite, as described in Equation 1.

$$\alpha_{cura} = 1 - \frac{\Delta H_{res}}{\Delta H_{total}} \quad (1)$$

Where ΔH_{res} is the residual enthalpy of the curing reaction, obtained by DSC, and ΔH_{total} is the total enthalpy of the curing reaction of the uncured material. In this regard, the degree of cure corresponds to the fraction of the curing reaction that has already occurred in the material, and is an important parameter for evaluating the crosslinking state of the polymeric matrix and its influence on the thermal and mechanical properties.

1.4 RESULTS AND DISCUSSION

1.4.1 Residual Thermal Behavior of Sisal/Epoxy Composites Subjected to Different Curing

Figure 3 presents the TG/DTG and DSC curves of the epoxy matrix and the sisal/epoxy [0°] composite, obtained from residual thermal analysis under different curing conditions.

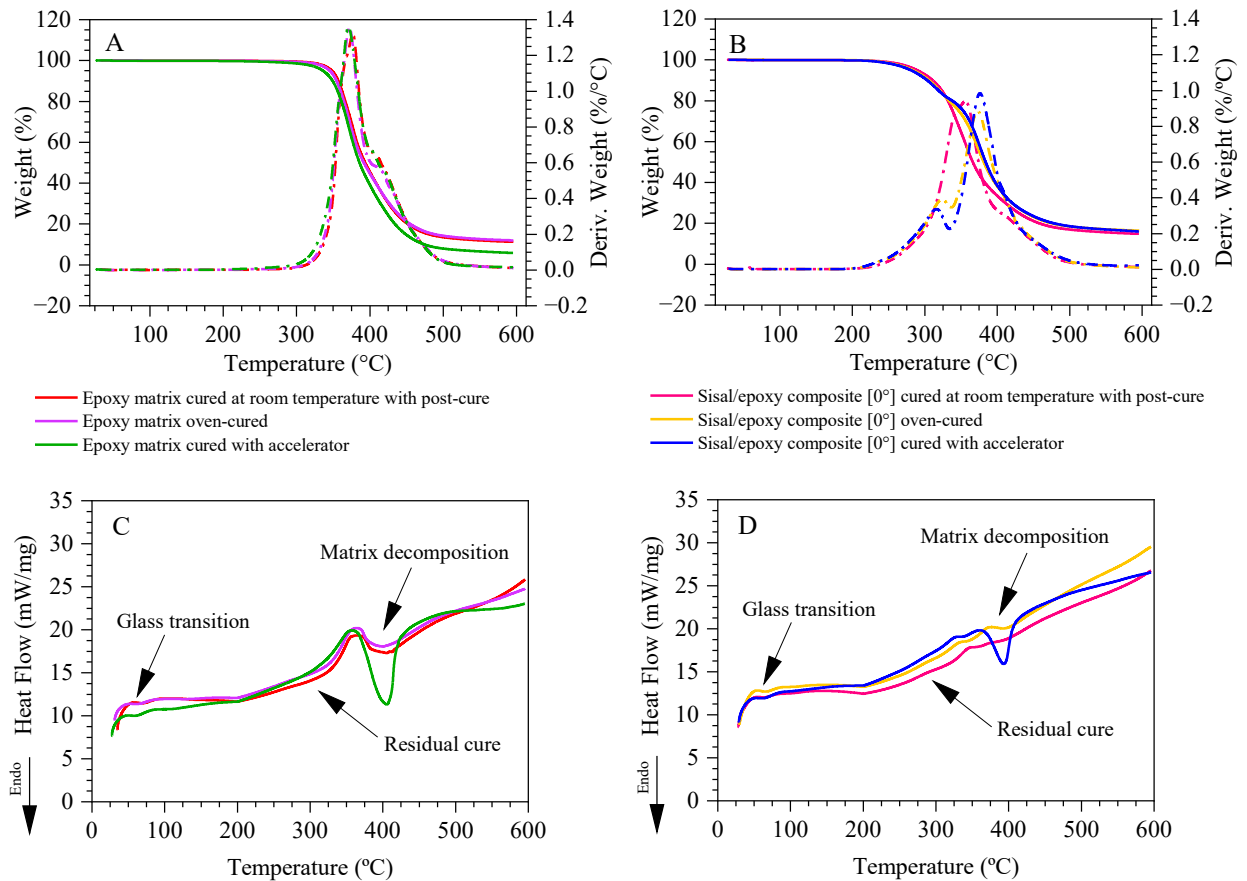


Figure 3 – TG/DTG curves of (A) the epoxy matrix and (B) the sisal/epoxy composite [0°], and DSC curves of (C) the epoxy matrix and (D) the sisal/epoxy composite [0°].

From the TG and DTG curves presented in Figures 3A and 3B, it is observed that, for both the epoxy matrix and the sisal/epoxy [0°] composite, no significant changes in the thermal profile were observed as a function of the different curing conditions applied to the matrix. However, when comparing the materials, the epoxy matrix exhibited greater thermal stability than the composite. This behavior is related to the single-phase nature of the matrix, whose thermal response depends exclusively on the polymer network formed during curing.

In the composite, the thermal behavior results from the interaction between the epoxy matrix and the sisal fiber. Due to the lignocellulosic nature of the fiber, whose constituents exhibit lower thermal stability than the polymeric matrix, decomposition is anticipated, resulting in reduced thermal stability of the system [66]. Furthermore, the composite's DTG curve exhibits at least two degradation peaks, reflecting the complexity of the decomposition process arising from the simultaneous contribution of the polymeric matrix and the sisal fiber constituents. Although the epoxy matrix also exhibits at least two peaks, indicating multi-step decomposition, these events occur within a narrower temperature range compared to those observed for the composite.

In addition, DSC analysis allows the identification of the thermal events associated with the observed transformations. For both the epoxy matrix (Figure 3C) and the sisal/epoxy composites (Figure 3D), at least three distinct thermal events are observed. The first is associated with the glass transition of the matrix, a phenomenon characteristic of amorphous and semicrystalline polymers, in which the material transitions from the glassy to the flexible state [67]. The second event can be attributed to residual matrix cure, resulting from crosslinking reactions that were not completed during processing. Finally, the third event corresponds to the thermal decomposition of the epoxy matrix, occurring within the same temperature range as the main mass loss observed in the TG/DTG curves (Figure 3A) [68].

Given these observations, it becomes pertinent to quantify the thermal parameters of the analyzed materials. Thus, Table 3 presents the thermal properties of the epoxy matrix and the sisal/epoxy composite under different curing conditions, considering the results obtained before and after the creep test. For comparative purposes, only the parameters related to the initial decomposition temperature (thermal stability) and the degree of cure of the system were highlighted.

The choice of these parameters is since thermal stability is directly related to the onset of the material's decomposition. Thus, its evaluation enables identification of potential changes in the system's thermal resistance resulting from creep loading. The degree of cure, in turn, is related to the extent of polymerization and crosslinking in the epoxy matrix, representing the fraction of reactive groups converted during curing.

Table 3 – Thermal stability and degree of cure of the epoxy matrix and sisal/epoxy [0°] composite under different curing conditions, before and after creep.

Samples / Property	Epoxy Matrix Before Creep			Epoxy Matrix After Creep		
	Room temperature with post-cure	Oven	Accelerator	Room temperature with post-cure	Oven	Accelerator
Thermal Stability (°C)	289	285	278	298	295	292
Degree of Cure (%)	95.9	92.7	92.3	98.2	98.7	99.2

Samples / Property	Sisal/Epoxy Composite [0°] Before Creep			Sisal/Epoxy Composite [0°] After Creep		
	Room temperature with post-cure	Oven	Accelerator	Room temperature with post-cure	Oven	Accelerator
Thermal Stability (°C)	132	137	177	212	215	220
Degree of Cure (%)	88.1	85.0	86.9	95.4	95.2	95.7

From the data presented in Table 3, it is observed that before creep, the epoxy matrix exhibited higher thermal stability than the sisal/epoxy [0°] composite across all evaluated curing conditions. This result confirms the higher thermal resistance of the matrix, as previously discussed, since its decomposition is associated exclusively with the polymeric network. In the case of the composite, the lower thermal stability values are related to the presence of sisal fiber, whose constituents exhibit lower thermal resistance and begin decomposing at lower temperatures than the matrix.

Furthermore, before the creep test, the degree of cure of the epoxy matrix was higher than that of the composite across all evaluated curing routes. The incorporation of the fiber into the system modifies the matrix crosslinking kinetics, as the presence of the fiber/matrix interface may restrict the mobility of polymer chains during curing, thereby limiting the progress of crosslinking reactions [69]. As a result, the composite tends to exhibit a lower degree of cure than the neat epoxy resin.

After the creep test, an increase in both thermal stability and degree of cure was observed for the epoxy matrix and the sisal/epoxy [0°] composite, regardless of the curing condition adopted. This increase in thermal stability occurs concomitantly with the increase in cure percentage. It may be associated with the continuation of crosslinking reactions that had not been fully completed, as well as possible structural rearrangements within the polymer network.

Even after creep, the epoxy matrix maintained thermal stability higher than that observed for the composite. Although the composite exhibited increased thermal stability and degree of cure, its thermal response remained influenced by the presence of the lignocellulosic phase. It is also observed that the increase in the degree of cure after creep was consistent for both materials, suggesting that mechanical loading did not compromise the thermal integrity of the system; rather, it may have contributed to a higher degree of crosslinking.

Regarding the influence of the curing conditions applied to the matrix, it is observed that prior to the creep test, variations in the thermal stability of the epoxy matrix were relatively minor across the evaluated curing routes. Room-temperature curing followed by post-curing exhibited thermal stability up to 289 °C. In comparison, oven curing reached 285 °C, and accelerator curing reached 278 °C, corresponding to a maximum difference of approximately 10 °C among the curing conditions analyzed.

After creep, the differences among the curing conditions became even less pronounced, with thermal stability values of 298 °C for room-temperature curing followed by post-curing, 295 °C for oven curing, and 292 °C for curing with an accelerator, establishing a variation of approximately 5 °C among the curing routes analyzed. Even with these small variations, room-temperature curing followed by post-curing exhibited the highest thermal stability values, both before and after creep. According to Dutra *et al.* [70], the additional post-cure step allows reactions that were not completed during the initial curing stage to proceed, resulting in an increased degree of crosslinking in the polymer network.

Furthermore, before creep testing, the highest degree of cure in the epoxy matrix was observed for room-temperature cure (95.9%), followed by post-cure (95.9%), oven cure (92.7%), and accelerator-cured (92.3%). After creep testing, the degree of cure increased under all evaluated conditions, reaching 98.2% for room-temperature cure followed by post-cure, 98.7% for oven cure, and 99.2% for cure with an accelerator. Despite the increase in

cure percentages, the values became very close to one another, attenuating the differences initially observed among the curing routes.

For the sisal/epoxy [0°] composite, a distinct behavior is observed regarding the influence of curing conditions on the material's thermal stability. Before creep, curing with an accelerator exhibited the highest thermal stability value (177 °C), exceeding oven curing (137 °C) by approximately 29.2%, and room-temperature curing followed by post-curing (132 °C) by approximately 34.1%. After creep, this trend was maintained, with the highest thermal stability observed for the sample cured with accelerator (220 °C), followed by oven curing (215 °C), room-temperature curing, and post-curing (212 °C). Despite these differences, the values remained close to one another, indicating similar thermal behavior across the evaluated curing conditions.

Regarding the degree of cure of the composite, before creep testing, room-temperature cure followed by post-cure exhibited the highest percentage (88.1%), followed by accelerator-cured (86.9%) and oven-cured (85.0%). After creep testing, the cure percentages increased, reaching 95.7% for accelerator-cured, 95.4% for room-temperature cure followed by post-cure, and 95.3% for oven cure. Despite the increase, the values became very close to one another, suggesting that creep loading contributed to the advancement of residual matrix curing, reducing the differences initially observed among the curing conditions.

1.4.2 Residual Mechanical Behavior of Sisal/Epoxy Composites Subjected to Different Curing Conditions

Figure 4 presents the stress-strain curves obtained from residual tensile testing after creep for the epoxy matrix and the sisal/epoxy composites, under different curing conditions and fiber orientations.

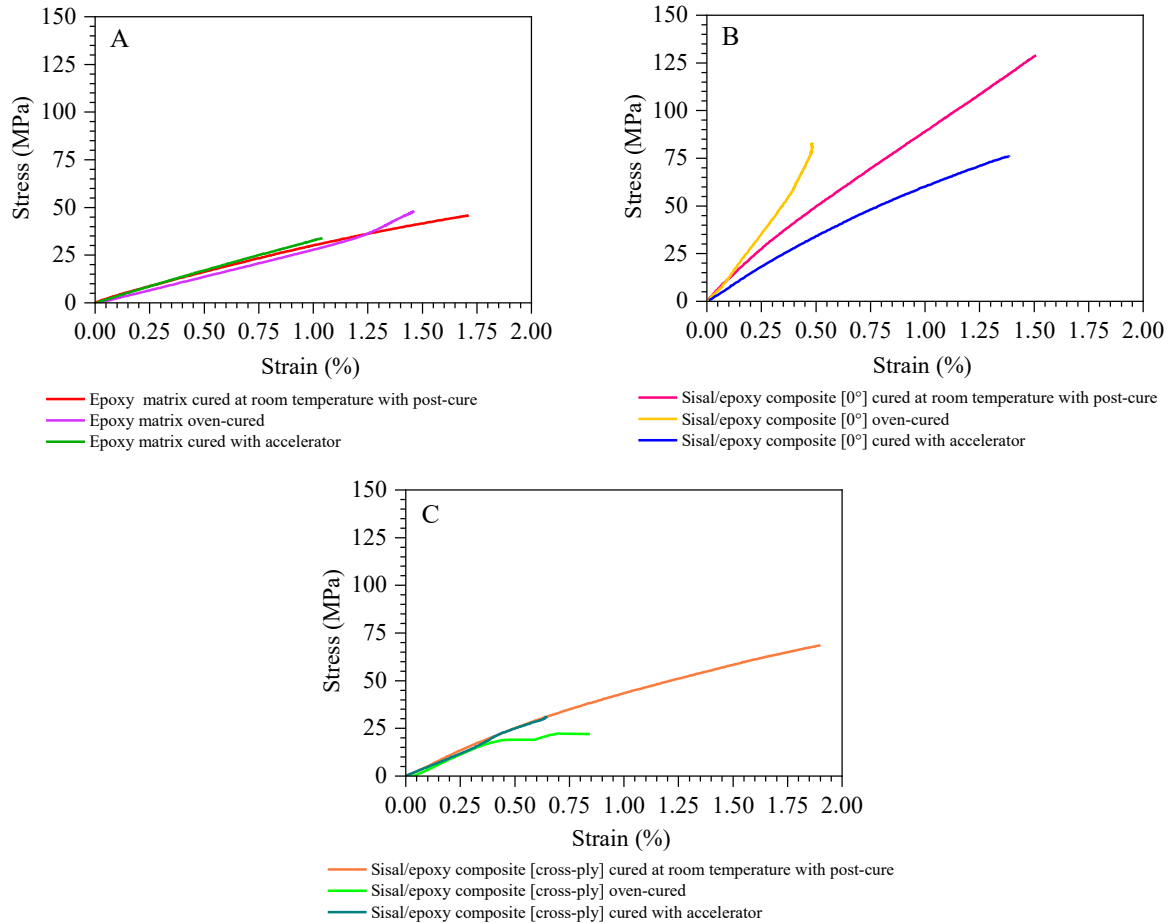


Figure 4 – Stress–strain curves of the epoxy matrix (A), the sisal/epoxy composite [0°] (B), and the sisal/epoxy composite [cross-ply] (C).

From the curves presented in Figure 4, it is observed that for all samples, stress increases continuously with strain, beginning with a linear region associated with the elastic response of the material. As strain increases, the curves exhibit changes in slope, indicating variations in stiffness throughout the loading process and differences in the stress levels sustained [71]. This behavior demonstrates that, although the materials exhibit the expected general trend in tensile tests, the evolution of the mechanical response under deformation differs between the epoxy matrix and the sisal/epoxy composites.

In the case of the epoxy matrix, a characteristic behavior of thermoset polymeric materials is observed, marked by a less stiff initial response and lower stress levels than those

of the composites. The lower slope of the stress-strain curves indicates a lower elastic modulus and, consequently, a limited capacity to resist deformation under tensile loading, since, in the absence of reinforcement, the polymeric matrix fully bears the applied stresses.

On the other hand, incorporating sisal natural fibers significantly increases the stiffness of the composites, as these fibers act as structural elements that bear and redistribute applied stresses. In composites, particularly those with fibers oriented in the loading direction, the initial elastic response is predominantly governed by the fibers, which are stiffer than the polymeric matrix. Under these conditions, the applied load is efficiently transferred from the matrix to the reinforcement, resulting in a more stable linear response.

A similar behavior was reported by Wu *et al.* [72], who investigated epoxy matrix composites reinforced with natural silk and flax fibers. The authors found that incorporating fibers significantly increased tensile strength compared to the neat epoxy matrix. The composite with the highest fiber content exhibited a tensile strength approximately 73% greater than that of the unreinforced matrix, a result attributed to the high mechanical load-bearing capacity provided by the natural fibers. These findings demonstrate the potential of natural fibers as reinforcing elements to improve the mechanical properties of polymeric composites significantly.

Given these observations, it is pertinent to systematically quantify the mechanical parameters of the analyzed materials. In this context, Table 4 presents the main properties obtained from the tensile tests of the epoxy resin and the sisal/epoxy composites. In addition to variations in curing conditions and fiber orientations, the table compiles the mechanical parameter results before and after the creep test. It is noted that the results for the condition before creep were extracted from the studies by Teixeira *et al.* [15], which present experimental tensile data for the materials in their initial state. For comparative purposes, the parameters related to elastic modulus, tensile strength, and strain at failure were considered.

Table 4 – Mechanical properties of the epoxy matrix and the sisal/epoxy composites under different curing conditions and fiber orientations, before and after creep testing.

Samples	Tensile Strength (MPa)		Strain (%)		Elastic Modulus (GPa)	
	Before Creep	After Creep	Before Creep	After Creep	Before Creep	After Creep
Epoxy matrix cured at room temperature with post-curing	41.54	45.94	1.44	1.71	2.92	3.47
Epoxy matrix cured at 100 °C/4 h in an oven	35.47	48.11	1.12	1.46	2.78	3.27
Epoxy matrix cured with accelerator	25.99	33.99	0.89	1.04	3.08	3.54
Sisal/epoxy [0°] cured at room temperature with post-curing	87.24	128.89	1.51	1.57	8.85	10.79
Sisal/epoxy [0°] cured at 100 °C/4 h in an oven	81.50	75.08	1.00	0.63	12.65	11.49
Sisal/epoxy [0°] cured with accelerator	71.61	60.47	1.24	0.71	12.37	7.84
Sisal/epoxy [cross-ply] cured at room temperature with post-curing	58.91	72.24	1.45	1.50	7.66	10.40
Sisal/epoxy [cross-ply] cured at 100 °C/4 h in an oven	26.30	22.34	0.84	0.57	6.34	5.53
Sisal/epoxy [cross-ply] cured with accelerator	36.23	24.20	0.70	0.53	6.03	4.72

Based on the results presented in Table 4, prolonged exposure to loading through the creep test tended to influence the mechanical behavior of both the epoxy matrix and the sisal/epoxy composites, with these effects depending on the curing condition and fiber orientation. Overall, the results suggest that creep may have promoted changes in the matrix and the fiber/matrix interface, as reflected in variations in strength, stiffness, and deformation capacity of the materials.

For the epoxy matrix, an increase in mechanical properties was observed after the creep test, regardless of the curing condition used. This behavior is consistent with the results of the thermal analyses, which showed increases in the degree of cure and thermal stability of the material after the test. Together, these results suggest a possible evolution of residual matrix curing over the experimental period, favoring the development of a more cohesive and stable polymeric structure. However, this behavior cannot be attributed solely to mechanical loading, as natural matrix aging during the 24 h test period may also have contributed to the observed changes.

Upon analyzing the curing conditions individually, the oven cure yielded the greatest gains in mechanical properties after creep, with increases of approximately 35.6% in tensile strength, 30.4% in strain at failure, and 17.6% in elastic modulus. Cure with accelerator presented increases of approximately 30.8% in tensile strength, 16.9% in strain at failure, and 14.9% in elastic modulus. In contrast, room temperature cure followed by post-cure exhibited the smallest gains, with increases of approximately 10.6% in tensile strength and 18.8% in both strain at failure and elastic modulus.

Regarding the sisal/epoxy composites, overall mechanical performance superior to that of the neat epoxy matrix is observed, both in terms of strength and stiffness. The fibers act as structural reinforcing elements, bearing a significant portion of the applied stresses and redistributing them throughout the polymeric matrix. Furthermore, the presence of lignocellulosic reinforcement increases system stiffness and enhances load-bearing capacity, particularly when the fibers are oriented in the direction of the applied stress [73].

When comparing laminate architectures, $[0^\circ]$ -oriented composites exhibited superior mechanical performance relative to cross-ply systems. The alignment of fibers parallel to the loading direction favors more efficient load transfer between the matrix and the reinforcement, resulting in higher strength and stiffness. In cross-ply laminates, a portion of the fibers is oriented transversely to the direction of the applied stress, which limits their effective contribution to load bearing and reduces the mechanical efficiency of the system [74].

Upon evaluating the effects of creep, distinct responses are observed depending on the curing conditions. In composites cured at room temperature and then post-cured, an increase in mechanical properties was observed after testing. The $[0^\circ]$ -oriented laminate exhibited a gain in tensile strength from 87.24 MPa to 128.89 MPa, corresponding to an increase of approximately 47.7%. Strain at failure and elastic modulus also increased, although to a lesser extent. For the cross-ply composite under the same curing conditions,

tensile strength increased from 58.91 MPa to 72.24 MPa, representing an increase of approximately 22.6%.

In contrast, the composites cured in an oven and with an accelerator exhibited reduced mechanical performance after the creep test, in both the $[0^\circ]$ configuration and the cross-ply arrangement. In these systems, a decrease in tensile strength and elastic modulus was observed, along with a loss of load-bearing capacity under prolonged loading. Unlike the epoxy matrix, whose behavior is predominantly governed by the polymeric network, the performance of the composites is strongly dependent on the integrity of the fiber/matrix interface.

Room-temperature cure, followed by post-cure, proceeds more gradually, allowing better accommodation of the matrix around the fibers and reduced development of residual stresses at the interface [75]. This condition preserves the integrity of fiber/matrix adhesion and promotes more stable load transfer throughout the creep test. On the other hand, oven-cure and accelerator-cure processes involve faster reaction kinetics and, in the case of oven-cure, greater thermal gradients, which may intensify the formation of internal stresses and discontinuities in the interfacial region. Under creep loading, these discontinuities facilitate damage accumulation at the fiber/matrix interface, compromising load transfer efficiency and reducing the overall mechanical performance of the composite.

1.5 CONCLUSIONS FROM CHAPTER I

Based on the results obtained, the analyzed materials exhibited changes in their thermal and mechanical properties after the creep test exposure period, with this response directly conditioned by the matrix curing state and the quality of the fiber/matrix interface.

From a thermal standpoint, the epoxy matrix exhibited higher thermal stability than the sisal/epoxy composite. Before creep, the matrix exhibited thermal stability ranging between 278 °C and 289 °C, while the composite recorded values between 132 °C and 177 °C. After creep, an increase in this parameter was observed for both materials, with the epoxy matrix exhibiting values between 292 °C and 298 °C and the composite between 212 °C and 220 °C. These results confirm the higher thermal resistance of the epoxy matrix and demonstrate that, although incorporating sisal fibers reduces the composite's thermal stability, the material maintains stable thermal behavior even under creep loading.

Regarding the curing conditions, the epoxy matrix exhibited slight variations in thermal stability, with differences of less than 10 °C before creep and less than 5 °C after creep; the best performance was observed for room-temperature curing, followed by post-curing. In contrast, for the composite, curing with an accelerator yielded the highest thermal stability, reaching 177 °C before creep and 220 °C after creep, surpassing the values obtained with the other curing routes.

With respect to the degree of cure, prior to creep testing, the epoxy matrix exhibited higher values, ranging from 92% to 95%, whereas the composite exhibited values between 84% and 88%. After creep loading, a significant increase in this parameter was observed in both materials, reaching values close to 98% for the matrix and 95% for the composite. These results suggest that creep loading acted as an activation mechanism for residual curing, promoting the advancement of polymer matrix crosslinking.

Regarding residual mechanical behavior, the materials' response after creep testing was nonuniform and strongly dependent on the matrix curing condition, fiber orientation, and fiber/matrix interfacial integrity. In general, the incorporation of sisal fibers promoted a significant increase in the stiffness and strength of the composites compared to the neat epoxy matrix, particularly in systems with fibers oriented in the loading direction ($[0^\circ]$), in which load transfer occurred more efficiently.

For the epoxy matrix, creep testing resulted in improved mechanical properties under all curing conditions evaluated, with oven cure presenting the most notable gains, including increases of 35.6% in tensile strength, 30.4% in strain at failure, and 17.6% in elastic

modulus. The results suggest that this behavior may be related to the progression of residual curing reactions under loading, thereby promoting a higher crosslink density in the polymeric matrix and, consequently, greater stiffness and load-bearing capacity.

On the other hand, in the sisal/epoxy composites, the creep response depended on the curing conditions and laminate architecture. In systems cured at room temperature followed by post-cure, an improvement in mechanical performance was verified after creep testing, with increases in tensile strength from 87.24 MPa to 128.89 MPa ($\approx 47.7\%$) in the $[0^\circ]$ laminate and from 58.91 MPa to 72.24 MPa ($\approx 22.6\%$) in the cross-ply arrangement. In contrast, composites cured in the oven and with an accelerator exhibited a reduction in mechanical properties after creep testing, which is associated with the accumulation of damage at the fiber/matrix interface.

Overall, the results suggest that the behavior of the analyzed materials under creep loading arises from a balance between residual matrix curing and mechanisms at the fiber/matrix interface. While creep may have favored the progression of crosslinking, reflected in improved thermal properties and, in some cases, improved mechanical performance, particularly in systems with better interfacial quality, it may also have contributed to mechanical degradation in conditions where the fiber/matrix interface proved less efficient.

CHAPTER II – ANALYSIS OF THE DYNAMIC-MECHANICAL PROPERTIES OF SISAL FIBER-REINFORCED COMPOSITES AND FIBER-METAL LAMINATES

2.1 INTRODUCTION

Under real operating conditions, polymer composites are widely used in components subjected to dynamic loading, in which cyclic stresses and temperature variations act simultaneously over time. Under these conditions, the mechanical response of these materials becomes more complex, as it involves the combined action of multiple deformation mechanisms associated with the multiphase nature of the system and interactions among its constituents [76].

From a mechanical standpoint, these materials exhibit viscoelastic behavior, characterized by simultaneous dependence on time, temperature, and loading frequency. This behavior results from the mobility of polymer chains and the interaction between the matrix and the reinforcement, directly influencing the capacity of the material to store and dissipate energy [55]. In this context, dynamic mechanical analysis stands out as an important tool for the characterization of these properties, enabling the determination of parameters that provide information on the stiffness and energy dissipation of the materials [77].

The incorporation of natural fibers into polymeric matrices significantly alters the dynamic-mechanical response, generally associated with increased stiffness and changes in dissipative mechanisms, due to matrix-reinforcement interactions and efficient stress transfer. However, factors such as fiber/matrix interfacial efficiency, fiber orientation, surface treatments, and processing conditions exert a direct influence on the final performance of the composite [78].

In addition to conventional composites, hybrid systems combining natural and synthetic fibers have been widely investigated as an alternative to reconcile mechanical performance and sustainability. Hybridization allows for the exploitation of synergies among different reinforcements, resulting in materials with higher stiffness and damping capacity [79]. In this context, fiber-metal laminates (FMLs), which combine metallic sheets with fiber-reinforced composites, also stand out, further expanding the application potential of these materials [80].

From a dynamic-mechanical standpoint, both natural fiber-reinforced composites and fiber-metal laminates exhibit responses strongly dependent on the constitutive characteristics of their components and the quality of the interfaces between phases [81].

The parameters obtained from DMA are widely used to evaluate stiffness, energy dissipation, and molecular relaxation mechanisms present in these systems. The analysis of these parameters enables an understanding not only of the viscoelastic behavior of the materials but also of the effects of hybridization and laminate architecture on the overall material performance.

Given this context, it is essential to systematically investigate the dynamic-mechanical behavior of sisal fiber-reinforced composites and fiber-metal laminates across different structural configurations and processing conditions. Understanding these aspects is essential for developing materials with optimized properties and reliable performance in applications subject to dynamic loading. Thus, this chapter aims to analyze the dynamic-mechanical properties of sisal fiber-reinforced composites and fiber-metal laminates using dynamic mechanical analysis, seeking to correlate the materials' viscoelastic response with their microstructural characteristics and manufacturing conditions.

2.2 LITERATURE REVIEW

2.2.1 Viscoelasticity and Dynamic Mechanical Analysis of Polymeric Materials

Polymeric materials exhibit intrinsically time-, temperature-, and strain-rate-dependent mechanical behavior, characterized by a viscoelastic response. This behavior results from the combination of the elastic properties typical of solids and the viscous properties of fluids. In practical terms, viscoelasticity manifests as the capacity of polymers to store energy (elastic component) and dissipate it as heat (viscous component) under cyclic or long-term mechanical loading [55].

From a molecular standpoint, viscoelasticity is related to the segmental mobility of polymer chains, which is influenced by the polymer's chemical structure, the degree of chain entanglement, the presence of crosslinking, and environmental conditions. Thus, at low temperatures or under high strain rates, polymers tend to respond in a stiffer, more elastic manner, whereas at elevated temperatures or under slow loading rates, the behavior becomes more viscous, with increased permanent deformation [82].

For the characterization of this behavior, dynamic mechanical analysis (DMA) tests are widely employed. DMA is a technique used for characterizing the viscoelastic behavior of polymers and composites, particularly when the aim is to understand how these materials respond to variations in temperature, frequency, and time [83]. In this test, the application of an oscillatory force allows the determination of parameters such as the storage modulus (E'), the loss modulus (E''), and the damping factor ($\tan \delta$), which are associated with the capacity of the material to store and dissipate energy [84].

During a DMA test, an oscillating force is applied to the sample and the material response to this excitation is measured. The applied force and the resulting deformation do not occur simultaneously; instead, there is a characteristic lag between them, represented by the phase angle δ [85,86]. This phase lag, illustrated in Figure 5A, reflects the coexistence of elastic and viscous components in the internal structure of the material.

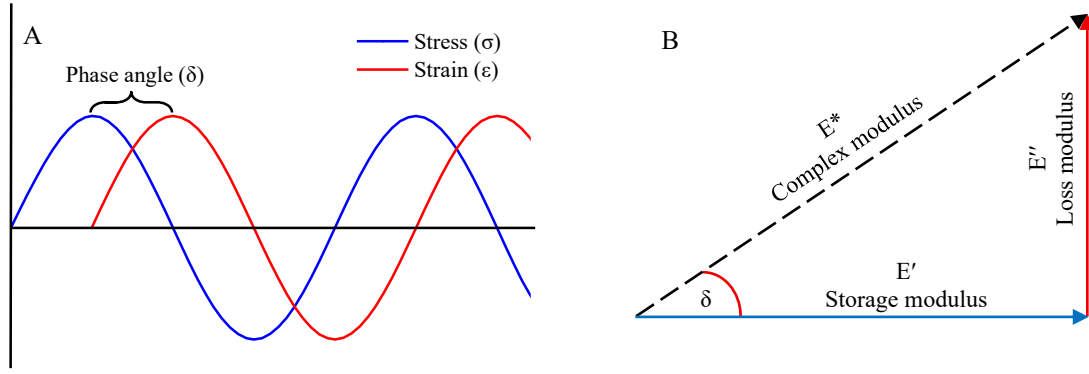


Figure 5 – Sinusoidal oscillation (A) and relationship between the storage modulus and the loss modulus (B). Adapted from Mulla *et al.* [83].

From this relationship, the complex modulus (E^*) emerges as the parameter representing the overall resistance of the material to deformation under dynamic loading. Unlike the Young's modulus obtained under static conditions, E^* describes the mechanical response to a sinusoidal excitation, simultaneously integrating the energy storage and dissipation mechanisms that occur during alternating loading [87]. Thus, E^* can be expressed by Equation 2, in which σ corresponds to the applied stress and ϵ to the resulting strain.

$$E^* = \frac{\sigma}{\epsilon} \quad (2)$$

Or alternatively in decomposed form, according to Equation 3:

$$E^* = E' + iE'' \quad (3)$$

As illustrated in Figure 5B, the complex modulus is composed of two variables: the real part, E' , and the imaginary part, iE'' . The storage modulus (E') corresponds to the elastic component of the mechanical response and indicates the amount of energy the material is capable of conserving in each oscillatory cycle. This quantity is directly associated with the dynamic stiffness of the system, such that materials with high E' values tend to exhibit greater resistance to deformation [83,87]. E' can be represented by Equation 4, in which δ represents the phase angle between the stress and strain signals.

$$E' = E^* \cos \delta \quad (4)$$

The E' response is sensitive to internal structural changes, such as segmental mobility, degree of crosslinking, reinforcement orientation, and volume fraction, as well as fiber/matrix interfacial efficiency [88]. In thermoset polymers, for example, increases in the

degree of cure or crosslink density generally increase E' . In composites, higher E' values are typically associated with good interfacial adhesion and efficient load transfer between the matrix and the fibers [89].

The loss modulus (E''), in turn, quantifies the energy dissipated as heat, characterizing the viscous response of the material [90]. This parameter is associated with internal friction mechanisms and molecular relaxation processes that occur during deformation, which promote irreversible energy losses in each oscillatory cycle. Thus, part of the applied mechanical energy is not recovered by the material and is converted into heat [91]. E'' can be defined according to Equation 5:

$$E'' = E' \tan \delta \quad (5)$$

Furthermore, the E'' peak is frequently used to identify specific relaxation processes, such as the glass transition (T_g) or secondary relaxations associated with the movement of side groups or small polymer chain segments. In these regions, energy dissipation reaches its maximum, increasing molecular mobility and intensifying internal friction and viscous deformation mechanisms [92]. In polymeric composites, both the intensity and position of this peak are influenced by the efficiency of the fiber/matrix interface, where more restrictive interfacial interactions limit segmental mobility and alter the dissipative profile of the material [93]. Figure 6 illustrates the relationship between E' and E'' , which can be understood from the behavior of a bouncing ball that stores and dissipates energy.

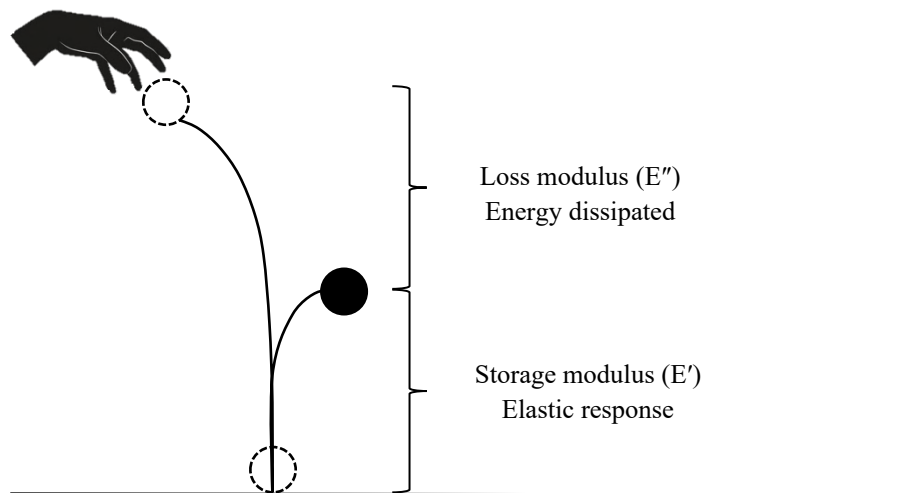


Figure 6 – Relationship between E' and E'' . Adapted from Mulla *et al.* [83].

The ratio between E'' and E' is expressed by the damping factor, or loss tangent ($\tan \delta$), as presented in Equation 6. This dimensionless parameter directly describes the balance between the elastic and viscous components of the mechanical response of the material [94].

Higher $\tan \delta$ values indicate that the viscous component predominates, meaning the material dissipates a significant fraction of mechanical energy as heat due to greater molecular mobility and internal friction. In contrast, lower $\tan \delta$ values indicate that the elastic response is dominant, characterizing a stiffer material that conserves a greater portion of energy during oscillatory deformation [95].

$$\tan \delta = \frac{E''}{E'} \quad (6)$$

In polymeric composites, the magnitude of $\tan \delta$ is also related to the efficiency of the fiber/matrix interface. When interfacial adhesion is effective, the mobility of polymer chains near the reinforcement surface becomes more restricted, reducing the friction and relaxation mechanisms responsible for energy dissipation [96]. Being sensitive to these internal structural changes, the $\tan \delta$ response also allows for the interpretation of thermal phenomena in the material, such as the peak observed in the curve, which corresponds to the point at which a significant increase in segmental mobility occurs, serving as an indicator of the glass transition temperature (T_g) of the material [97,98].

The glass transition temperature (T_g) corresponds to the range in which an amorphous polymer, or the amorphous fraction of a semicrystalline polymer, undergoes a significant change in mechanical behavior, transitioning from a rigid and brittle state to a more flexible, rubber-elastic state. Unlike the melting temperature (T_m), T_g is characterized by a gradual increase in the segmental mobility of polymer chains, without structural changes occurring in the molecular network [99]. In DMA, T_g can be determined using several methods, such as the point of maximum energy dissipation ($\tan \delta$ peak), the loss modulus (E'') peak, or the inflection point observed in the storage modulus (E') curve [100].

For the glass transition to occur, polymer chains must initiate cooperative motions, which are established when the material possesses sufficient energy to activate its relaxation mechanisms. The minimum energy required for these molecular segments to overcome the energy barriers is called the activation energy. In DMA, when the test is conducted at different frequencies, this energy can be estimated through thermorheological models, such as the Arrhenius or Williams–Landel–Ferry models [101]. At higher frequencies, the material is subjected to loading conditions that demand faster chain responses, requiring greater energy for cooperative motions to occur. As a consequence, a shift of T_g to higher temperatures is observed. Similarly, at lower frequencies, since more time is available for molecular accommodation, T_g tends to shift to lower temperatures [102].

The influence of T_g extends beyond relaxation mechanisms, being directly related to the degree of matrix crosslinking. In thermoset systems, an increase in crosslink density often leads to a higher T_g , as the three-dimensional network formed by these bonds reduces the freedom of movement of polymer chains [81,103]. This effect is not limited to the transition point, becoming even more evident in the flexible region.

Figure 7 illustrates this behavior by comparing two materials with distinct crosslink densities: one highly crosslinked and one with reduced crosslinking. It is observed that the more crosslinked material presents significantly higher E' and E'' values in the flexible region, indicating a stiffer and more constrained structure. The less crosslinked material, in contrast, exhibits lower moduli, consistent with a less dense network and reduced restriction to segmental motion [103].

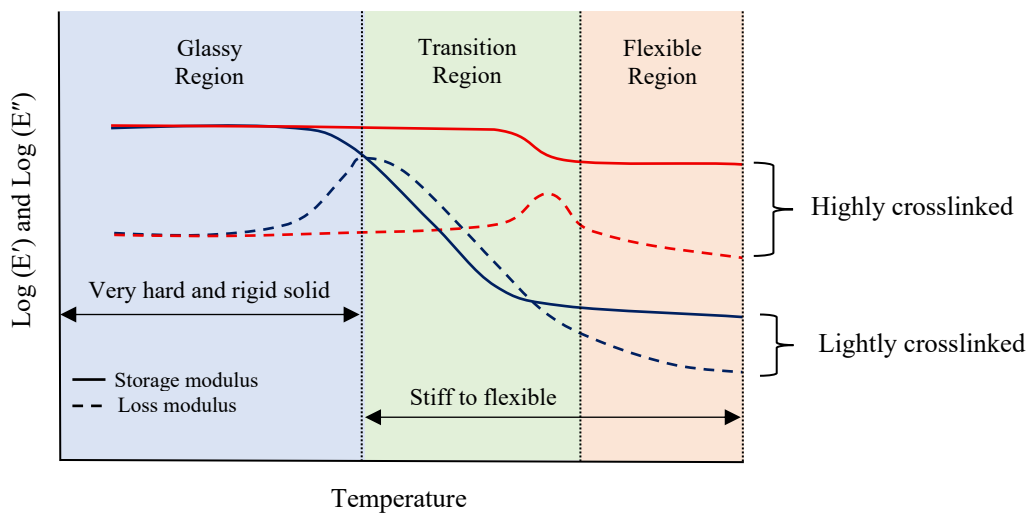


Figure 7 – Representation of E' and E'' of composites with high and low crosslink densities.
Adapted from Haris *et al.* [103]

Furthermore, T_g determines the service conditions of the material. Flexible elastomers, such as polyisobutylene and polyisoprene, are used in the rubbery state, that is, above T_g . In this range, polymer chains exhibit greater freedom of movement, imparting high flexibility and good deformation capacity to the material. Polymers such as polyvinyl chloride and polystyrene, on the other hand, are used in their glassy state, below T_g . Under this condition, chain mobility is limited by the molecular structure itself, resulting in stiffer and dimensionally stable materials [81,96].

2.2.2 Dynamic-Mechanical Properties of Natural Fiber-Reinforced Polymer Composites

The use of natural fibers as reinforcement in polymer composites has established itself as an alternative to improve the mechanical performance, and particularly the dynamic-mechanical properties of these materials. In addition to being a sustainable and economically viable alternative, these fibers exhibit a high strength-to-weight ratio and good specific stiffness, characteristics that favor the formation of lighter, mechanically stable structures when incorporated into the polymeric matrix [104,105].

From a viscoelastic standpoint, the presence of reinforcement significantly alters the dynamic response of the material compared to the neat matrix. The incorporation of fibers tends to increase the stiffness of the system, since the restriction imposed on the movement of polymer chains reduces molecular mobility and promotes more efficient load transfer throughout the composite [106]. As a result, the materials exhibit lower viscosity and a greater predominance of elastic response compared to the neat polymeric matrix.

The combination of these effects yields a material with higher mechanical efficiency and improved thermal stability, highlighting the potential of natural fibers to modulate dynamic behavior. The literature has consistently reported such improvements, demonstrating that even in systems where the fibers do not achieve ideal adhesion levels, their contribution to viscoelastic performance remains significant, particularly compared with the more limited behavior of the polymeric matrix [103].

In the studies by Silva *et al.* [107] on kenaf/epoxy composites, it was observed that incorporating fibers enhanced the dynamic-mechanical performance of the material relative to the neat resin. Both the storage and loss moduli were higher, particularly in the composite with the highest fiber content. Similarly, Oliveira *et al.* [108] observed the same behavior in epoxy resin composites reinforced with fique fabrics. In this study, the authors concluded that the composite exhibited superior properties to those of the neat epoxy resin, demonstrating the positive effect of fibrous reinforcement on the viscoelastic response of the material.

Despite these advantages, the use of natural fibers in polymeric matrices still faces challenges inherent to these fibers. Being composed of cellulose, hemicellulose, and lignin, which are highly hydroxylated, natural fibers exhibit a hydrophilic character and a high tendency toward moisture absorption. This characteristic, combined with the presence of waxes, pectins, and other extractives, compromises their interaction with the polymeric

matrix [109]. As a result, interfacial adhesion is limited, leading to the formation of discontinuity zones that compromise the overall performance of the composite [110].

The presence of these discontinuities directly affects how the material responds to dynamic loading. When the fiber/matrix interface is inefficient, load transfer is incomplete, which intensifies segmental mobility in the matrix. Under these conditions, the capacity of the fiber to restrain deformation decreases, causing the mechanical response of the material to depend more heavily on the polymeric matrix. Thus, the composite tends to exhibit reduced stiffness, higher internal energy dissipation, and greater sensitivity to variations in temperature and frequency [111].

These challenges, however, can be mitigated through surface treatments applied to the fibers, whose primary objective is to modify their surface characteristics, reduce hydrophilicity, and improve chemical interaction with the polymeric matrix [112]. Treatments such as alkalization, silanization, and acetylation, as well as physical processes such as plasma and corona treatments, remove amorphous fractions and introduce functional groups that promote more effective adhesion between the matrix and the reinforcement [113].

In this context, several studies have investigated the effects of these treatments on the viscoelastic properties of composites. Islam *et al.* [114] concluded, in their study of jute/epoxy composites, that the incorporation of fibers increased the stiffness of the material, an effect even more pronounced when treated fibers were used. Similar results were reported by Lekrine *et al.* [115], who studied polylactic acid (PLA) biocomposites with hybrid reinforcement of biomass and palm fibers. The authors found that treatment of the fibers in sodium bicarbonate (NaHCO_3) solution increased the elastic and viscous properties of the material, resulting in E' and E'' values higher than those of the untreated samples.

In addition to the effects associated with the chemical composition of the fibers and the improvements promoted by surface treatments, the dynamic-mechanical performance of composites also depends on how the reinforcement is organized within the matrix. When fibers are longitudinally aligned (parallel to the loading direction), the reinforcement is most effectively utilized, resulting in greater load-transfer efficiency, higher stiffness, and improved thermal stability. In this configuration, the segmental motion of the matrix is significantly restricted, reducing energy dissipation during dynamic loading [116].

On the other hand, when fibers are arranged transversely or randomly, the composite makes less use of its reinforcing potential. In these orientations, the capacity of the fibers to contribute to stiffness is partially lost, increasing the influence of the matrix on the

mechanical response. Under these conditions, stiffness is reduced and energy dissipation increases, due to greater segmental mobility in the matrix and lower load-transfer efficiency between the fiber and the matrix [117].

Doddi *et al.* [118] investigated the effect of fiber orientation of pineapple leaf fibers used as reinforcement in epoxy resin composites. The study considered five orientation angles (0° , 30° , 45° , 60° , and 90°) relative to the direction of the applied load. The authors observed that the best dynamic-mechanical properties were obtained when the fibers were aligned at 0° , that is, parallel to the loading direction. Similarly, Hassan *et al.* [119] evaluated shape memory polymer composites reinforced with flax fibers, considering three orientations (0° , 45° , and 90°) relative to the loading direction. The results showed that laminates with fibers aligned at 0° exhibited greater stiffness and better thermal stability, while those oriented at 90° yielded the poorest performance.

In general, incorporating natural fibers into polymeric matrices effectively improves the dynamic-mechanical performance of composite materials, particularly when their orientation, arrangement, and surface treatment are carefully controlled. The combination of these factors contributes to the balance between stiffness and damping capacity, resulting in lighter, more stable materials with more efficient mechanical behavior.

2.2.3 Dynamic-Mechanical Properties of Hybrid Composites Reinforced with Natural and Synthetic Fibers

Hybrid composites are materials formed by combining two or more types of reinforcements within a single polymeric matrix, which may be arranged in distinct layers or distributed simultaneously throughout the laminate. This strategy allows the individual characteristics of each fiber type to be synergistically combined, producing systems with superior performance compared to composites reinforced with a single fiber type [120].

In this context, the hybridization of natural and synthetic fibers has attracted increasing attention, not only for the potential to enhance the mechanical performance of these materials but also for offering more environmentally sustainable solutions. Synthetic fibers, such as glass and carbon, provide high strength and structural stability, while natural fibers are distinguished by their low density, lower cost, and renewable origin. The combination of these characteristics enables the development of lighter, more efficient, and environmentally friendlier materials [121].

The mechanical response of hybrid composites is strongly influenced by interactions between different types of reinforcements and by the stress distribution throughout the

laminate. Synthetic fibers tend to increase the storage modulus, promoting greater stiffness and dimensional stability [122]. Conversely, the presence of natural fibers introduces greater structural heterogeneity into the composite, owing to their lignocellulosic composition and complex internal morphology. This characteristic influences the deformation mechanisms and alters the viscoelastic response of the polymeric matrix [123].

Beyond stiffness, the dissipative behavior of these materials is also affected. The lower interfacial compatibility of natural fibers with the matrix, combined with the complexity of their internal structure, contributes to increases in the loss modulus and damping factor. This effect arises from additional energy-dissipation processes, such as internal friction at the fiber/matrix interface and localized deformation within the fibrous structure, thereby rendering hybrid composites more efficient at attenuating vibrations than non-hybrid systems [124].

Hybridization also influences the dynamic-thermal behavior of composites, particularly with regard to the molecular mobility of the matrix. The incorporation of natural fibers may shift the glass transition temperature or broaden the transition range, depending on the interface quality and the degree of constraint imposed on the polymer chains [125]. These variations are critical for defining the operating conditions in applications that are simultaneously subjected to mechanical loading and thermal fluctuations. Table 5 below compiles recent studies that investigated the dynamic-mechanical properties of polymeric composites with hybrid natural and synthetic fiber reinforcement, addressing the influence of hybridization on the performance of these materials.

Table 5 – Recent studies on hybrid composites of natural and synthetic fibers in thermosetting matrices.

Matrix	Hybrid reinforcement	Reference
Polyester	Jute/glass	Das, S. C., <i>et al.</i> (2021) [126]
Epoxy	Flax/glass/carbon	Mabrouk, O. M., <i>et al.</i> (2024) [127]
Epoxy	<i>Calotropis procera</i> /glass	Raghu M. J. and Govardhan Goud (2022). [128]
Polyester	Sisal/glass	Ornaghi, H. L., <i>et al.</i> (2021) [129]
Epoxy	Flax/glass/hemp	Karademir, C., <i>et al.</i> (2025) [130]
Polyester	Palm/carbon	Madhan, M. G. K., Rajeshkumar, G., and Ravikumar, P. (2023) [131]

Das *et al.* [126] investigated the effect of jute/glass fiber hybridization as reinforcement in polyester composites. Overall, the hybrid composites exhibited higher stiffness and lower $\tan \delta$ peaks compared to the non-hybridized materials. Furthermore, the

hybrid systems presented glass transition temperature (T_g) values superior to those of composites reinforced with a single fiber type.

Mabrouk *et al.* [127] examined the hybridization of flax fibers with glass and carbon fibers, as well as the flax/carbon/glass combination, in epoxy matrix composites. The results generally indicated that the flax/carbon system exhibited the best mechanical performance, with higher storage and loss moduli than the other composites analyzed.

Raghu M. J. and Govardhan Goud [128] evaluated the replacement of 5, 10, 15, and 20 wt% of *Calotropis procera* fibers with glass fibers. The results demonstrated that incorporating glass fibers increased both the storage and loss moduli. In addition, the $\tan \delta$ peaks became narrower and less intense, with a predominance of the elastic response over the viscous one.

Ornaghi *et al.* [129] conducted a study on sisal/glass hybrid composites, varying the fiber proportion in the composite. The results showed that hybridization improved the dynamic-mechanical properties relative to the neat polyester matrix, with the best performance observed in the composite with the highest glass fiber content.

Karademir *et al.* [130] investigated the dynamic-mechanical properties of hybrid composites reinforced with flax/glass and hemp/glass fibers. The authors found that incorporating glass fibers enhanced the viscoelastic properties of the materials, as reflected primarily in increased stiffness and improved dynamic response of the system.

Madhan, Rajeshkumar and Ravikumar [131] investigated the influence of hybridization on the mechanical properties of polyester composites reinforced with palm and carbon fibers. The results demonstrated that the addition of carbon fiber to the palm fiber-reinforced system yielded significant improvements in mechanical performance, particularly in the viscoelastic behavior, with increases in E' and E'' values relative to the neat matrix and to composites containing only a single fiber type.

As observed in non-hybrid composites, the mechanical performance of hybrid composites is strongly influenced by several factors, among which the stacking sequence of the reinforcement layers is particularly noteworthy. The relative positioning of the different layers determines the distribution and transfer of stresses throughout the laminate, directly affecting the stiffness and internal energy dissipation of the material [132].

When the outer layers of the laminate consist of rigid fibers, such as synthetic fibers, the composite exhibits greater stiffness and stability, as these layers bear most of the applied load and restrict matrix deformation. Conversely, when the outer layers are composed of more flexible fibers, such as natural ones, the matrix has greater freedom of movement,

which increases energy dissipation and confers higher damping capacity to the composite [133].

In the studies by Selver *et al.* [134], the influence of stacking sequence on the viscoelastic behavior of flax/glass and jute/glass hybrid composites was analyzed. The results showed that the layer configuration plays a determining role in the viscoelastic behavior of the materials: laminates with glass fibers in the outer layers exhibited greater stiffness, whereas those with natural fibers, such as flax or jute, in the outer layers displayed higher damping capacity.

Complementarily, Arquier *et al.* [135] evaluated hybrid composites reinforced with flax and carbon fibers and observed a similar trend. The authors found that composites with carbon fibers in the outer layers presented the highest E' values, whereas those with flax fibers in the outer layers exhibited considerably lower E' values. This behavior was attributed to the higher intrinsic stiffness of carbon fibers and their greater resistance to stresses during testing.

Overall, the studies presented demonstrate that the dynamic-mechanical performance of hybrid composites results from the interaction between the nature of the reinforcements and their arrangement within the laminate. The presence of synthetic fibers primarily increases stiffness and structural stability, while natural fibers play a more significant role in energy dissipation, conferring greater damping capacity on the material.

In this regard, hybridization enables the strategic tuning of the viscoelastic behavior of composites, thereby balancing seemingly antagonistic properties, such as stiffness and energy dissipation. Furthermore, the stacking sequence constitutes a fundamental parameter in this process, as the relative positioning of the layers influences the efficiency of load transfer and the degree of constraint on the mobility of the polymeric matrix chains.

2.3 MATERIALS AND METHODS

2.3.1 Sisal/Epoxy Composites Subjected to Different Curing Conditions

Composite materials used in this study were produced in a previous work developed by Teixeira *et al.* [15]. Overall, the composites were manufactured by compression molding, using Araldite® LY 5052 epoxy resin and Aradur® 5052 hardener at a ratio of 100:38, with a 50% fiber volume fraction of sisal fibers. The fibers were previously treated in a 5% (w/v) aqueous NaOH solution, at a solution-to-fiber ratio of 10:1, at 50 °C for 2 h under manual stirring. Table 6 presents the main characteristics of the materials analyzed.

Table 6 – Dimensions of the materials subjected to the DMA test, including the epoxy matrix and sisal/epoxy composites under different curing conditions and fiber orientations.

Material	Curing Condition	Dimensions (mm)		
		Length	Width	Thickness
Epoxy Matrix	Cured at room temperature with post-curing	51.2	9.7	2.5
	Cured at 100 °C/4 h in an oven	50.7	9.6	2.9
	Cured with accelerator	50.2	9.7	2.2
Sisal/Epoxy [0°]	Cured at room temperature with post-curing	50.9	10.4	2.6
	Cured at 100 °C/4 h in an oven	48.1	10.7	2.9
	Cured with accelerator	51.0	10.6	2.4
Sisal/Epoxy [45°]	Cured at room temperature with post-curing	51.2	11.9	2.8
	Cured at 100 °C/4 h in an oven	50.9	11.0	3.6
	Cured with accelerator	50.8	11.3	2.3
Sisal/Epoxy [cross-ply]	Cured at room temperature with post-curing	50.6	10.3	3.0
	Cured at 100 °C/4 h in an oven	50.9	10.7	2.7
	Cured with accelerator	51.2	10.6	3.5

Unidirectional composites were manufactured with 0° and 45° orientations, as well as in the cross-ply configuration [0°/90°/90°/0°]. The samples were subjected to three distinct curing routes: (i) curing at room temperature for 24 h, followed by post-curing in an oven at 120 °C for 1 h; (ii) curing in an oven at 100 °C for 4 h; and (iii) curing at room temperature with the addition of an accelerator at a 100:38:1 ratio, without subsequent post-curing. For comparison, unreinforced epoxy matrix samples were also produced and subjected to the same curing conditions as the composites.

2.3.2 Fiber-Metal Laminates with Hybrid Natural and Synthetic Fiber Reinforcement

The fiber-metal laminates used in this study were produced in a previous work carried out by Libera Junior [16]. Overall, unidirectional sisal/epoxy, carbon/epoxy, and glass/epoxy prepregs were manufactured, all with a fiber volume fraction of 50%. For the production of the FMLs (Fiber-Metal Laminates), 2024-T3 aeronautical aluminum alloy sheets with a thickness of 0.5 mm were used.

Three groups of unidirectional laminates were produced. The first group consisted of FRP (Fiber-Reinforced Polymer) laminates, obtained from the carbon, glass, and sisal fiber prepregs. The second group comprised non-hybrid FMLs, formed by combining aluminum sheets with a single FRP composite type. The third group, in turn, consisted of hybrid FMLs that combined the aluminum sheet with two types of FRP composites in alternating arrangements of synthetic- and natural-fiber FRPs. The composition and layer distribution within the FRP and FML laminates are presented in Figure 8 and Table 7.

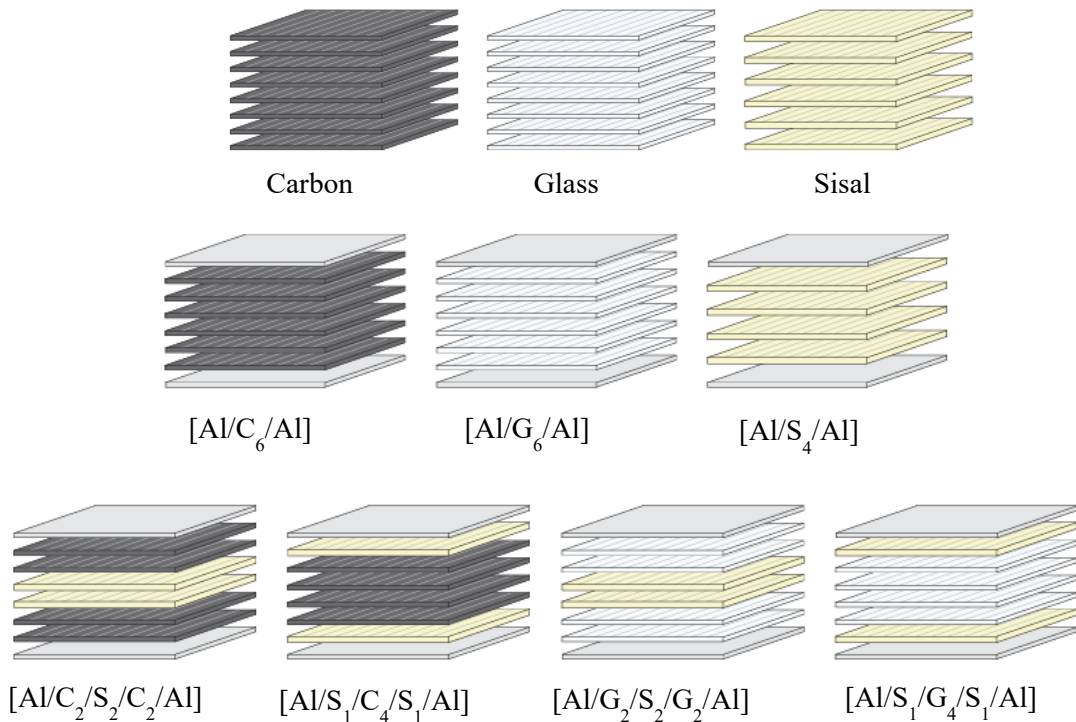


Figure 8 – Schematic representation of the architecture and stacking sequence of the FRP and FML laminates. Adapted from Libera Junior [16].

Table 7 – Dimensions of materials subjected to DMA testing, composition, and layer distribution of FRP and FML laminates.

Group	Laminate and Stacking Sequence	Number of Layers				Dimensions (mm)		
		Al	C	G	Al	Length	Width	Thickness
FRP	Carbon	-	8	-	-	50.2	12.1	2.2
	Glass	-	-	8	-	50.6	12.1	2.6
	Sisal	-	-	-	6	50.0	11.5	2.8
Non-hybrid FMLs	[Al/C ₆ /Al]	2	6	-	-	50.3	11.7	3.3
	[Al/G ₆ /Al]	2	-	6	-	50.6	10.8	3.8
	[Al/S ₄ /Al]	2	-	-	4	51.0	11.6	3.1
Hybrid FMLs	[Al/C ₂ /S ₂ /C ₂ /Al]	2	4	-	2	50.7	11.2	3.3
	[Al/S ₁ /C ₄ /S ₁ /Al]	2	4	-	2	50.6	11.4	3.6
	[Al/G ₂ /S ₂ /G ₂ /Al]	2	-	4	2	51.0	12.1	3.4
	[Al/S ₁ /G ₄ /S ₁ /Al]	2	-	4	2	51.8	11.1	3.6

The nomenclature adopted considered the number of layers in the laminate (8, 6, 4, or 2) and the constituent material of each layer (Al – aluminum, C – carbon, G – glass, or S – sisal). For example, the laminate [Al/C₆/Al] represents a non-hybrid FML composed of six carbon fiber layers.

2.3.3 Dynamic Mechanical Analysis (DMA)

The materials were characterized by dynamic mechanical analysis (DMA) using a NETZSCH model 242 E Artemis instrument. The tests were performed on rectangular specimens with approximate dimensions of (50 x 10 x 2.5) mm, employing a three-point bending fixture, as illustrated in Figure 9.

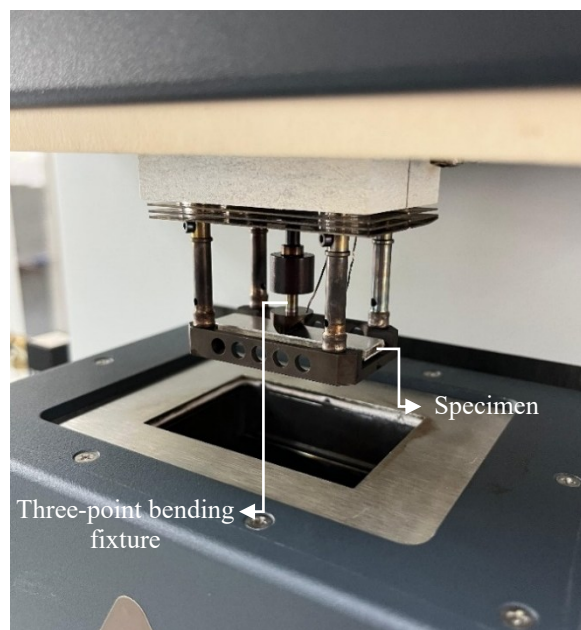


Figure 9 – Experimental setup for dynamic mechanical analysis (DMA) testing of the composites.

The analysis was performed over a temperature range of 35 °C to 220 °C, with a constant heating rate of 2 °C/min. During the tests, three distinct frequencies were applied: 0.1 Hz, 1 Hz, and 10 Hz. The activation energy (ΔE) was determined using the Arrhenius equation (Equation 7 [136]), which relates the dynamic loading frequency to the material's glass transition temperature.

$$\ln f = \ln A - \frac{\Delta E}{RT_g} \quad (7)$$

In this equation, f is the frequency used in the test, A is a constant factor related to the frequency, ΔE is the activation energy extracted from the slope of the $\ln f$ versus $1/T_g$ plot, and R is the universal gas constant (8.314 J/mol·K). Additionally, the DMA test allows evaluation of the reinforcement's influence on the mechanical response of the composites, as fiber incorporation significantly alters the material's stiffness and the load-transfer mechanisms between phases.

Based on this, the effect of reinforcement on the E' values of the sisal/epoxy composites was analyzed by determining the parameters C and A . The coefficient C is associated with reinforcement effectiveness and quantifies the contribution of the fibers to the increase in material stiffness. This parameter is obtained from the ratio between the E' values of the composite and the matrix, and can be calculated as indicated in Equation 8 [137]:

$$C = \frac{\left(\frac{E'_g}{E'_r} \right)_{composite}}{\left(\frac{E'_g}{E'_r} \right)_{matrix}} \quad (8)$$

where E'_g and E'_r are the storage moduli in the glassy region (at 50 °C) and the flexible region (at 150 °C), respectively. The reinforcement efficiency was also evaluated using the adhesion factor (A), which accounts for both E' and E'' , since $\tan \delta$ is implicitly included in the analysis. The adhesion factor A can be calculated according to Equation 9 [137]:

$$A = \frac{1}{1 - V_f} * \frac{\tan \delta_c}{\tan \delta_m} \quad (9)$$

where V_f is the fiber volume fraction (0.50 [15]), and $\tan \delta_c$ and $\tan \delta_m$ represent the damping values of the composite and the matrix, respectively.

2.4 RESULTS AND DISCUSSION

2.4.1 Dynamic-Mechanical Behavior of Sisal/Epoxy Composites Subjected to Different Curing Conditions

Figure 10 presents the E' and $\tan \delta$ curves as a function of temperature, obtained by DMA for the neat epoxy matrix and for the sisal/epoxy composites.

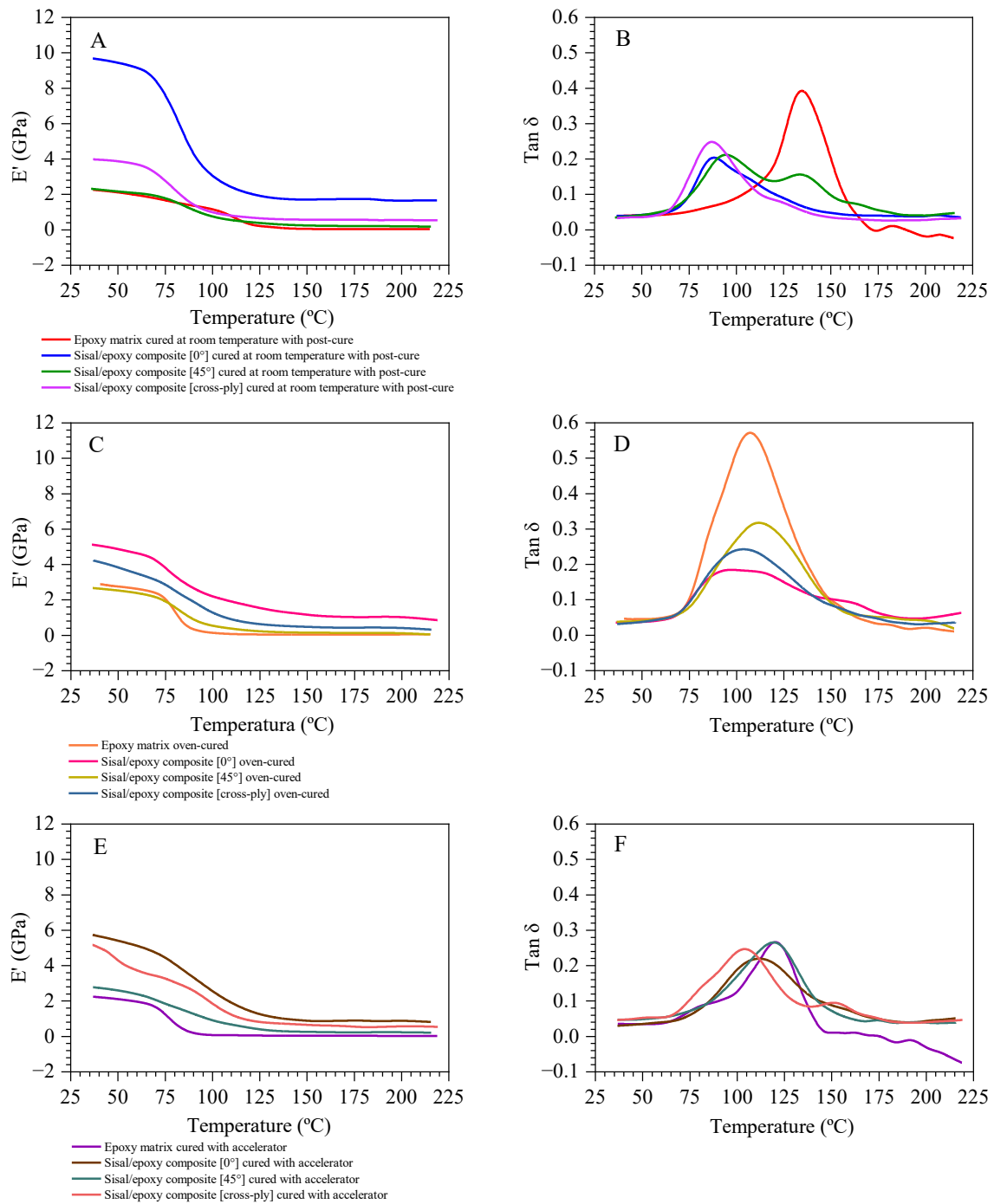


Figure 10 – Viscoelastic properties as a function of temperature, obtained at a frequency of 1 Hz: storage modulus (E') and loss factor ($\tan \delta$) for materials cured at room temperature with post-curing (A–B), oven-cured (C–D), and accelerator-cured (E–F).

From the curves presented in Figures 10A, 10C, and 10E, a characteristic behavior of viscoelastic polymeric materials is observed. In all samples, a reduction in E' with increasing temperature is verified. According to Jawaid *et al.* [138], this decrease is directly associated with the progressive activation of segmental mobility of the polymer chains, which intensifies as the matrix approaches the transition region.

At lower temperatures, the material remains in the glassy state, with the polymer chains rigidly immobilized, resulting in high E' values. However, as the temperature increases, the polymer chains begin to move more freely, reducing the material stiffness and, consequently, decreasing E' . According to Haris *et al.* [103], this behavior reflects the transition from the glassy to the flexible phase, which occurs as the temperature approaches the glass transition, at which point the viscoelastic properties of the material become predominant.

When comparing the epoxy matrix with the sisal fiber-reinforced composites, it is observed that, across the entire temperature range, the composites exhibit higher E' values than the matrix. This behavior confirms the role of the fibers as reinforcing elements that, upon incorporation into the matrix, restrict molecular mobility and increase system stiffness. At lower temperatures, for instance, the composites exhibited E' values more than twice those of the epoxy matrix.

In addition to increasing stiffness in the glassy state, the fibers also contribute to the mechanical stability of the material. While the epoxy matrix exhibits an abrupt drop in E' in the transition region, the composites maintain higher stiffness levels, retarding the loss of mechanical properties. In the flexible state, for example, the epoxy matrix presents very low E' values, approaching zero. In contrast, the composites retain higher stiffness levels due to the fibers, which prevent complete relaxation of the polymer network and maintain elevated E' values even in a highly damped regime.

Regarding the curves shown in Figures 10B, 10D, and 10F, the $\tan \delta$ values of the composites are lower than those of the epoxy matrix across all analyzed samples. This behavior indicates that the presence of fibers reduces the segmental mobility of the polymer chains, due to the more efficient transfer of stresses from the matrix to the reinforcement through the interface. In the epoxy matrix, the relaxation process depends exclusively on the amorphous molecular chains, resulting in greater energy dissipation. According to Ornaghi *et al.* [139], reduced $\tan \delta$ values indicate a more elastic character of the material. They are directly related to the quality of interfacial adhesion, which restricts molecular displacement

in the contact regions between the matrix and the fibers, thereby significantly affecting the energy dissipation capacity of the system.

As noted by Rahman [56], this effect can be explained by the fact that, in composites with good fiber-matrix interaction, the cooperative motions of the polymer chains are limited, reducing internal friction and, consequently, the amplitude of the $\tan \delta$ peak. Thus, the more effective the stress transfer and chain anchoring at the interface, the lower the dissipated energy. Conversely, in systems with poor interfacial adhesion, increased molecular mobility leads to higher $\tan \delta$ values.

In this context, to quantify the parameters derived from the DMA curves, Table 8 presents the main results of the dynamic mechanical analysis. Based on these results, the different curing conditions directly influenced the dynamic-mechanical properties of the analyzed materials. For the epoxy matrix, oven curing yielded the highest $\tan \delta$ value (0.57), corresponding to an increase of approximately 46% relative to the resin cured at room temperature, followed by post-cure ($\tan \delta = 0.39$), and approximately 111% compared to the accelerator-cured resin ($\tan \delta = 0.27$). This behavior indicates that, although oven curing promotes the formation of a more homogeneous and rigid three-dimensional network, regions with greater segmental mobility near the T_g persist.

For the composites, the $\tan \delta$ values were very similar across all curing conditions. However, the 45°- oriented composites exhibited slightly higher $\tan \delta$ values than the other fiber orientations, with the highest value recorded under oven curing ($\tan \delta = 0.32$). This behavior is associated with the lower structural efficiency of the fibers in this orientation, which renders the mechanical response of the composite more matrix-dependent. In this configuration, the matrix retains greater freedom of deformation, favoring energy dissipation and resulting in higher $\tan \delta$ values than in composites oriented at 0° or in cross-ply arrangements, in which the fibers are aligned, fully or partially, with the loading direction.

Regarding the T_g results, the highest value for the neat epoxy matrix was obtained for the sample cured at room temperature followed by post-cure (134.73 °C), followed by the accelerator-cured sample (120.16 °C), and finally the oven-cured sample (107.21 °C). The higher T_g observed in the post-cured sample results directly from the post-cure step, during which the polymerization reactions are completed, promoting additional crosslinking of the polymer network. In this step, partially entangled chains become interconnected, restricting segmental mobility. Consequently, higher temperatures are required for these chains to reorganize or undergo significant motion.

Table 8 – Viscoelastic properties of the epoxy matrix and sisal/epoxy composites.

Curing Condition	Material	T _g (°C) Tan δ	T _g (°C) E'' _{max}	Tan δ	E' _{max} (GPa)	E'' _{max} (GPa)	ΔE (KJ/mol)
Room temperature with post-curing	Epoxy Matrix	134.73	103.69	0.39	2.26	0.11	482.07
	Sisal/epoxy composite [0°]	88.18	83.67	0.20	9.68	1.10	395.84
	Sisal/epoxy composite [45°]	94.51	86.54	0.21	2.32	0.22	368.54
	Sisal/epoxy composite [cross-ply]	87.08	81.66	0.25	3.98	0.48	331.26
Oven 100 °C / 4 h	Epoxy Matrix	107.21	78.44	0.57	2.29	0.23	271.51
	Sisal/epoxy composite [0°]	97.22	86.67	0.18	5.13	0.48	316.03
	Sisal/epoxy composite [45°]	111.86	84.43	0.32	2.67	0.19	310.59
	Sisal/epoxy composite [cross-ply]	103.86	88.75	0.24	4.23	0.39	270.90
Accelerator	Epoxy Matrix	120.16	72.11	0.27	2.25	0.39	412.62
	Sisal/epoxy composite [0°]	111.70	100.19	0.22	5.73	0.49	273.50
	Sisal/epoxy composite [45°]	119.31	108.77	0.27	2.78	0.16	371.39
	Sisal/epoxy composite [cross-ply]	104.03	94.21	0.25	5.18	0.48	355.20

For the composites, the observed behavior was opposite. Across all fiber orientations, room-temperature curing followed by post-cure yielded the lowest T_g values, followed by oven curing, and finally accelerator curing. However, the E' response was found to be strongly dependent on fiber orientation and on the fiber-matrix interaction, indicating that, unlike the matrix, the stiffness of the composites is not governed exclusively by the segmental mobility of the matrix, but also by the efficiency of the reinforcing phase in constraining the imposed deformations.

In the 0° composite, where the fibers are aligned parallel to the loading direction, stress transfer occurs directly and efficiently, maximizing the structural contribution of the reinforcement. In this configuration, the highest E' value (9.68 GPa) was obtained for the sample cured at room temperature, followed by post-cure, although this condition yielded the lowest T_g (88.18 °C). This result can be explained by the combined effect of fiber alignment and the two-step polymerization process. The fiber contribution ensured increased stiffness, as the fibers play a predominant role in sustaining the applied loads. On the other hand, the post-cure step, while promoting additional crosslinking reactions, also introduced heterogeneity into the matrix, generating regions with varying degrees of molecular constraint.

For the 45° and cross-ply composites, the situation was distinct. In these configurations, the fiber contribution is less efficient, and the matrix and fiber/matrix interface assume a predominant role in the mechanical performance. Under room-temperature curing followed by post-cure, the E' values were reduced: 2.32 GPa for the 45° composite and 3.98 GPa for the cross-ply composite. These results demonstrate that, in these orientations, stress transfer from the matrix to the fibers is limited, particularly in the 45° composite, whose stiffness approached that of the epoxy matrix, confirming its greater matrix dependence. The cross-ply composite exhibited intermediate stiffness, attributed to the presence of 0° plies, which partially improved stress-transfer efficiency and helped sustain the applied loads.

Regarding thermal stability, as in the 0° composite, room-temperature curing followed by post-cure resulted in lower T_g values than those obtained under other curing regimes. For the 45° and cross-ply arrangements, T_g reached 94.51 °C and 87.08 °C, respectively. This behavior can be explained by the influence of fiber orientation on the distribution of internal stresses during curing. In arrangements where the fibers are not fully aligned with the loading direction, their structural contribution is reduced, making the material response more matrix-dependent. Under these conditions, the formation of heterogeneous regions arising from the two-step polymerization tends to be less pronounced, preserving greater uniformity of the polymer network and, consequently, yielding relatively higher T_g values compared to the 0° composite.

Under oven curing, an increase in T_g was observed for all fiber orientations, indicating higher thermal stability of the matrix. In the composites oriented at 45° and in the cross-ply arrangement, where stiffness is strongly dependent on the matrix and the fiber/matrix interface, this increase in T_g was directly reflected in a rise in the storage

modulus, with increases of 15.1% for the 45°-oriented composite (2.67 GPa) and 6.2% for the cross-ply arrangement (4.23 GPa) relative to the samples cured at room temperature followed by post-curing. These results show that the more stable matrix provided greater internal cohesion and better coupling with the fibers, favoring more efficient stress transfer. In the 0°- oriented composite, however, the behavior was distinct. Despite the increase in T_g (97.22 °C), the storage modulus decreased by approximately 47% compared to the sample cured at room temperature and then post-cured. This demonstrates that, for this orientation, the overall stiffness is governed by the contribution of the fibers, aligned with the loading direction.

Accelerator curing yielded the highest T_g values across all fiber orientations, indicating that the matrix exhibited greater thermal stability than the other curing routes. The use of an accelerator increases the reaction rate, promoting the faster formation of a relatively homogeneous polymer network. This greater uniformity more effectively restricts segmental motion in the matrix chains, thereby raising the T_g even without achieving the maximum degree of crosslinking. In the 45° and cross-ply composites, the increased matrix constraint and improved fiber-matrix coupling resulted in higher E' values (2.78 GPa and 5.18 GPa, respectively) than those observed under the other curing conditions.

In the 0° composite, although the T_g was significantly higher (111.70 °C), the storage modulus (5.73 GPa) was lower than that obtained under room-temperature curing followed by post-cure, yet still higher than that recorded under oven curing. These results indicate that, despite the matrix exhibiting greater thermal stability, the use of an accelerator may have promoted the formation of heterogeneous regions or minor interfacial defects, reducing the efficiency of stress transfer from the matrix to the fibers. As a result, even with a more thermally stable and partially crosslinked matrix, the composite stiffness did not reach the maximum values observed under room-temperature curing followed by post-cure.

As a complement to the preceding analysis, it is relevant to evaluate parameters that quantify reinforcement efficiency and fiber/matrix interfacial quality. Although the previous results demonstrated the influence of curing conditions and fiber orientation on segmental mobility and composite stiffness, the dynamic-mechanical performance analysis can be further deepened by incorporating indicators of reinforcement efficiency and efficacy. In this regard, coefficients C and A , presented in Table 9, enable an objective assessment of load transfer efficiency and interfacial behavior across different structural arrangements and curing regimes.

Table 9 – Reinforcement efficiency coefficient (C) and adhesion factor (A) for sisal/epoxy composites under different curing conditions and fiber orientations.

Curing Condition	Material	E'_g (GPa)	E'_r (GPa)	C	A
Room temperature with post-curing	Epoxy matrix	2.11	0.05	–	–
	Sisal/epoxy composite [0°]	9.43	1.71	0.138	1.026
	Sisal/epoxy composite [45°]	2.16	0.25	0.220	1.077
	Sisal/epoxy composite [cross-ply]	3.87	0.58	0.168	1.282
Oven 100 °C / 4 h	Epoxy matrix	2.75	0.05	–	–
	Sisal/epoxy composite [0°]	4.86	1.16	0.068	0.316
	Sisal/epoxy composite [45°]	2.53	0.15	0.269	0.561
	Sisal/epoxy composite [cross-ply]	3.82	0.48	0.130	0.421
Accelerator	Epoxy matrix	2.10	0.04	–	–
	Sisal/epoxy composite [0°]	5.40	0.86	0.112	0.815
	Sisal/epoxy composite [45°]	2.59	0.26	0.183	1.00
	Sisal/epoxy composite [cross-ply]	4.27	0.65	0.120	0.926

From Table 9, it is observed that fiber orientation significantly influences the efficiency and efficacy of the reinforcement in the composite. Laminates with fibers oriented at 0° presented the lowest values of both coefficient C and adhesion factor A , regardless of the curing condition. The reduction in coefficient C indicates greater load-transfer efficiency and higher overall system stiffness, while lower A values suggest a more effective interface for transmitting stresses along the fibers. In this configuration, the fibers are aligned with the loading direction, which favors the predominance of normal stresses at the interface and reduces the contribution of shear stresses. As noted by Ornaghi *et al.* [137], fiber/matrix

adhesion tends to increase as the fiber orientation angle decreases, reaching its maximum when the fibers are aligned with the loading direction.

Conversely, laminates with fibers oriented at 45° exhibited the highest values of both the coefficient C and the adhesion factor A . In this configuration, part of the applied load is borne by the matrix, whose modulus is lower than that of the fibers, resulting in a less efficient mechanical response. Furthermore, the 45° fiber angle relative to the loading axis promotes the development of shear stresses in the matrix and at the fiber/matrix interface, reducing the ability of the system to efficiently transfer stresses along the fibers.

The cross-ply composites exhibited intermediate values for both the coefficient C and the adhesion factor A . In this arrangement, the 0° plies improved load transfer and increased system stiffness, while the 90° plies introduced interlaminar stresses and stress-concentration regions. As a result, the overall performance of the material was superior to that of the 45° laminates, yet below the maximum efficiency observed in the fully aligned 0° laminates.

With respect to the curing conditions, the oven-cured composites generally presented the lowest values of both C and A . This behavior, however, is not directly associated with higher stiffness values or a greater degree of crosslinking, but rather with a more gradual E' transition over the temperature range, which reduces the difference between E'_g and E'_r . In the 0° composite, for example, the difference between the glassy and rubbery moduli was approximately 76%, whereas under post-cure conditions it was approximately 82%. Although this condition did not yield higher E'_g values, which were approximately 48% lower than those of the post-cured 0° laminate, it indicates that high-temperature polymerization promotes a more stable polymer network, with a narrower relaxation time distribution and a smoother thermal transition.

In the accelerator-cured composites, the C and A values fell within an intermediate range. Under this condition, the E'_g values, in some cases, exceeded those obtained under oven curing, although they remained approximately 43% lower than those observed under post-cure for the 0° laminate, for example. Nevertheless, the relative stiffness drop remained at around 83%, close to the post-cure condition, indicating that this curing route produces a less uniform polymer network than oven curing but is still more stable than the post-cure condition.

The composites cured at room temperature, followed by post-cure, presented the highest values of both C and A , evidencing a more abrupt glassy-to-flexible transition and a more pronounced relative difference between E'_g and E'_r . Although this curing route provides

a substantial increase in initial stiffness, notably the E'_g of 9.43 GPa in the 0° laminate, it also reveals greater heterogeneity in the polymer network. This behavior arises from the two-step polymerization process, in which the initial room-temperature cure is followed by an oven post-cure, which significantly increases the crosslink density but does not ensure the same level of structural uniformity as full oven curing.

These variations in reinforcement efficiency and interfacial quality are directly reflected in the homogeneity of the polymer system, which can be further assessed through Cole-Cole diagrams. Based on E' and E'' values, these diagrams can be constructed to analyze the distribution of molecular relaxation times and evaluate the homogeneity of the polymer system.

According to Chee *et al.* [140], the shape of the diagram provides important information about the nature of the system, since homogeneous polymeric materials tend to exhibit well-defined semicircular curves, while deviations from the ideal semicircle indicate structural heterogeneity, frequently associated with variations in the relaxation time distribution or with the quality of the fiber/matrix interface. Figure 11 presents the Cole-Cole diagrams of the epoxy matrix and the sisal/epoxy composites under the different curing conditions and fiber orientations.

According to the Cole-Cole diagrams, the composite curves do not form perfect semicircles, indicating that the system exhibits intrinsic heterogeneities and the existence of multiple relaxation mechanisms. According to Arulmurugan *et al.* [141], these heterogeneities may result from the multiphase nature of the material, involving the presence of fiber/matrix interfaces, variations in fiber distribution and impregnation, and local differences in the crosslink density of the epoxy matrix.

In the composites with fibers oriented at 0° , regardless of the curing condition, narrower semicircles are observed, suggesting greater stiffness and a more abrupt glass transition (see Figure 10). According to Ornaghi *et al.* [137], this behavior is attributed to the alignment of the fibers in the loading direction, which promotes stress transfer and confers greater elastic energy storage capacity prior to dissipation. Furthermore, the relationship between E' and E'' in these laminates is more uniform, suggesting a more efficient interaction between the polymeric matrix and the fibers.

As the fiber orientation angle increases (45° and cross-ply), the plots exhibit broader semicircles, indicating a mechanical response increasingly influenced by the polymeric matrix. In these configurations, the drop in E' throughout the transition region occurs more gradually (see Figure 10), evidencing a lower contribution of the fibrous reinforcement to

the overall stiffness and greater participation of the matrix in energy dissipation. Thus, the broadening of the semicircles simultaneously reflects both the greater dispersion of relaxation mechanisms and the dominant role of the polymeric phase in fiber orientations that are not aligned with the loading axis.

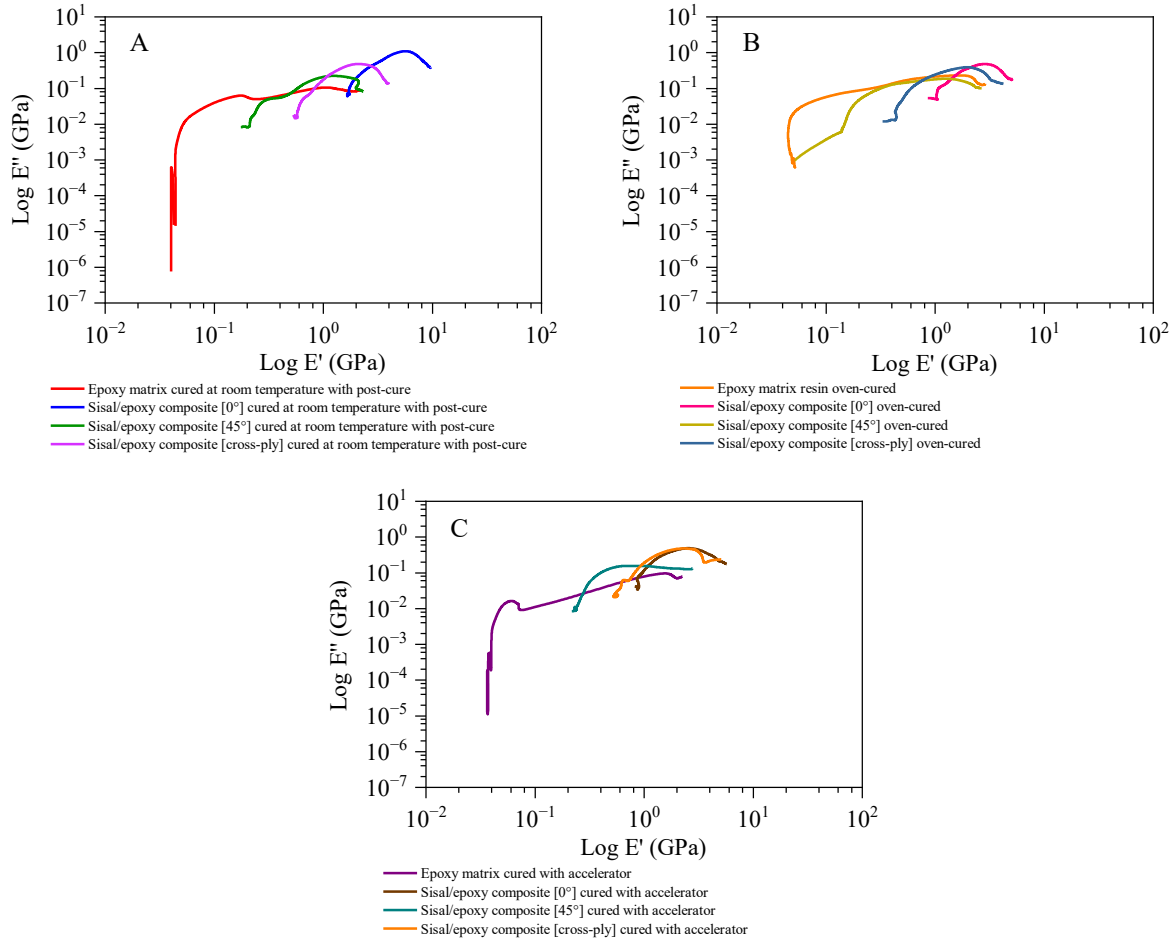


Figure 11 – Cole-Cole diagrams for the epoxy matrix and the sisal/epoxy composite, cured at room temperature with post-curing (A), in an oven (B), and with an accelerator (C).

In addition to fiber orientation, the curing conditions play an important role in the viscoelastic response of these materials. Oven curing, for example, yields more regular arcs closer to a semicircular shape, indicating a narrower dispersion of relaxation times and a more homogeneous polymer structure. This behavior is associated with the curing process conducted at elevated temperatures, a condition that ensures more stable and controlled reaction kinetics.

On the other hand, the materials subjected to room-temperature curing followed by post-curing exhibit a shift of the arcs toward higher E' values, indicating greater stiffness and elastic energy storage capacity (see Table 8). However, the irregular shape of the arcs suggests greater dispersion in relaxation times. This is attributed to the first curing stage,

which occurs more slowly, favoring the formation of regions with different degrees of crosslinking. Post-curing in an oven at high temperatures consolidates the polymer network, partially homogenizing these regions.

Curing with an accelerator, in turn, exhibited behavior similar to that observed for room-temperature curing followed by post-curing. The arcs displayed irregular shapes, indicating that although the accelerated reaction favors rapid development of the polymer network and formation of a sufficiently rigid three-dimensional structure (see Table 8), some regions still exhibit distinct degrees of crosslinking, resulting in a less homogeneous distribution of relaxation times.

2.4.2 Dynamic-Mechanical Behavior of Fiber-Metal Laminates with Hybrid Sisal and Synthetic Fiber Reinforcement

Figure 12 presents the E' and $\tan \delta$ curves as a function of temperature, obtained by DMA for the different laminate groups.

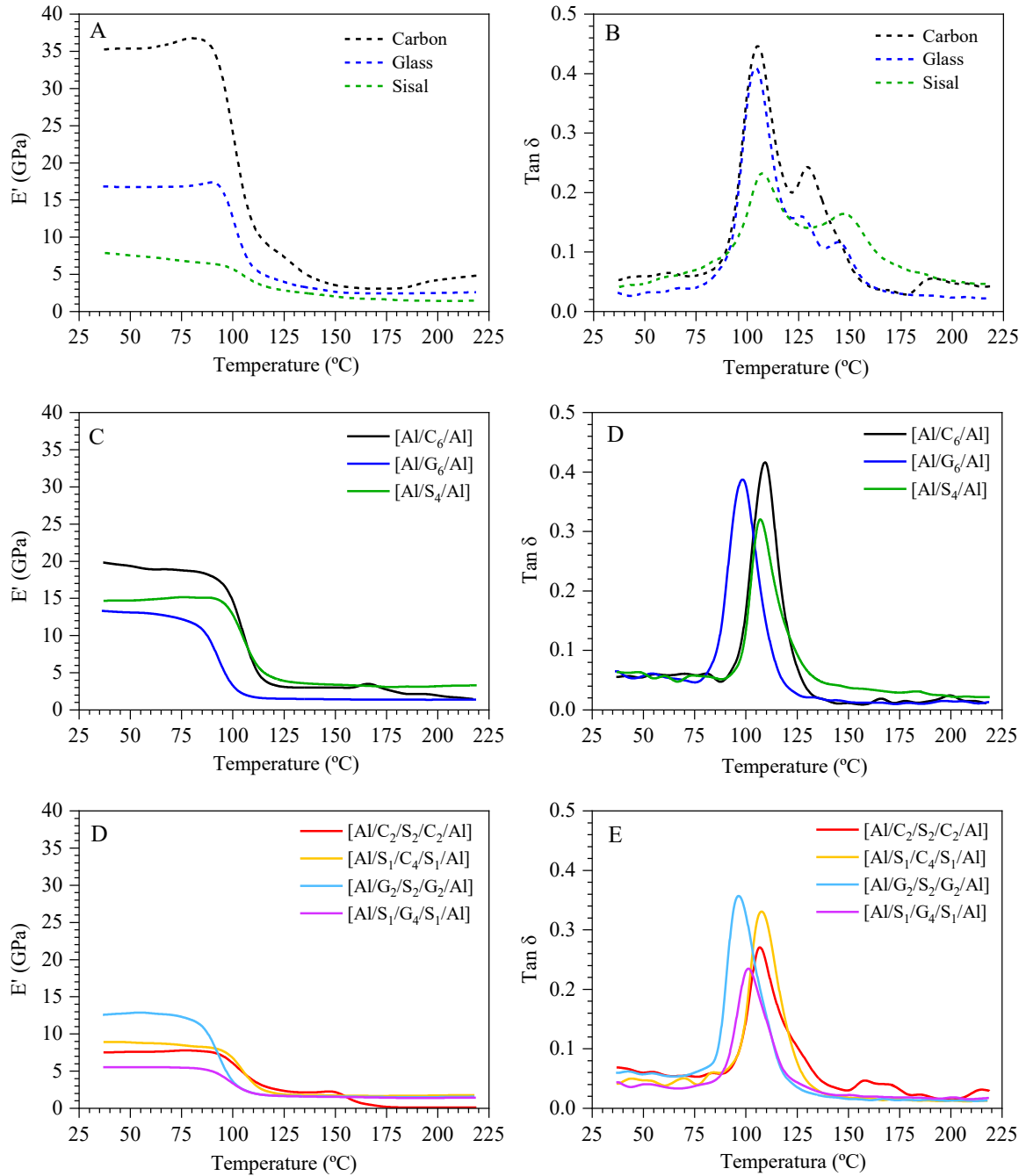


Figure 12 – Viscoelastic properties as a function of temperature, obtained at a frequency of 1 Hz: storage modulus (E') and loss factor ($\tan \delta$) for FRP composites (A–B), non-hybrid FMLs (C–D), and hybrid FMLs (E–F).

From the curves presented in Figures 12A, 12C, and 12E, the typical behavior of polymeric materials was observed, characterized by high stiffness at low temperatures and a progressive reduction in E' as temperature increases. However, beyond this general trend, some samples exhibited an additional phenomenon: an E' overshoot located near the transition region. In dynamic mechanical analysis, the overshoot manifests as a transient increase in E' that precedes the matrix glass transition. According to Menard [142], this behavior can be explained by different physical mechanisms, all associated with local matrix relaxation processes and the redistribution of internal stresses within the material.

Studies by Wu *et al.* [143], indicate that the difference between the thermal expansion coefficients of the matrix and the reinforcement causes the cure shrinkage of the matrix to be partially constrained by the reinforcement during processing, resulting in the development of residual internal stresses. These stresses remain stored within the material, creating regions with different levels of molecular constraint. Upon heating, these stresses are released, triggering a momentary recovery of stiffness. This behavior does not represent an improvement in material properties, but rather a transient response associated with molecular reorganization induced by the release of internal stresses.

In Fiber Reinforced Polymers (FRPs), the overshoot exhibited distinct behavior. The phenomenon was observed in systems reinforced with synthetic fibers but was absent in the natural fiber-reinforced material. This difference is associated with the intrinsic stiffness of the fibers. Synthetic fibers, such as carbon and glass, possess an elastic modulus higher than that of the polymeric matrix, which enables them to impose a more intense constraint on both matrix shrinkage during curing and subsequent molecular motion. This promotes the accumulation of internal stresses at the interface, which, upon release, result in a transient recovery of stiffness. In the case of sisal fibers, whose modulus is significantly lower than that of synthetic fibers, the ability to constrain the matrix's molecular motion is reduced, resulting in lower residual stress accumulation.

Another relevant aspect concerns the quality of the fiber/matrix interface. In FRPs composed of synthetic fibers, this interface tends to be more uniform and exhibit greater load-transfer efficiency. This interaction promotes the formation of cohesive regions with high molecular constraint, in which stress accumulates and is released in a pronounced manner. In FRPs containing sisal fibers, the interface is more heterogeneous, owing to the structural variability and surface irregularities intrinsic to natural fibers. This heterogeneity reduces the efficiency of interfacial adhesion, promoting a more gradual stress dissipation.

In non-hybrid Fiber Metal Laminates (FMLs), the observed behavior was opposite to that of the FRPs. In this case, the combination of synthetic fibers with metallic layers significantly reduced the occurrence of the overshoot. The aluminum, owing to its high elastic modulus, acted as a mechanical barrier to matrix expansion, limiting local variations in molecular mobility. As a result, the redistribution of internal stresses occurred gradually and homogeneously, preventing localized releases capable of inducing transient increases in E' . In these laminates, the matrix relaxed continuously up to the glass transition, without exhibiting abrupt stiffness fluctuations.

Regarding the hybrid FMLs, the occurrence of the overshoot was directly associated with the distribution of sisal fibers within the composite. When these fibers were positioned at the core of the laminate, they acted within a more constrained region, surrounded by high-stiffness layers composed of synthetic fibers and metallic plies. This structural arrangement created a mechanical barrier that significantly restricted the motion of the sisal fibers and the polymeric matrix. Unlike configurations in which sisal was located in less confined regions, at the core it was unable to efficiently accommodate the deformations and stresses generated during processing. Consequently, a significant portion of these stresses remained stored within the composite. Thus, upon heating, the confinement state prevented the gradual release of stresses, resulting instead in a more concentrated stress relief.

On the other hand, when sisal was positioned at the outer layers of the laminate, the composite behavior changed significantly. In this configuration, the sisal fibers were subject to considerably lower constraints, as they were not confined to the rigid core of the laminate. This additional freedom allowed the surrounding polymeric matrix to accommodate more uniformly during processing, preventing the localized accumulation of internal stresses. As a result, stress redistribution occurred continuously and efficiently, promoting the gradual dissipation of stored energy and significantly reducing the occurrence of transient stiffness peaks.

Based on the observations discussed above, it is relevant to quantify the parameters derived from the DMA curves to enable comparative analysis among the different types of laminates evaluated. In this context, Table 10 compiles the main results of the dynamic mechanical analysis, enabling a more in-depth assessment of the viscoelastic behavior of the materials as a function of the laminate architecture.

Table 10 – Viscoelastic properties of the different laminate groups.

Group	Laminate	T _g (°C) Tan δ	T _g (°C) E'' _{máx}	Tan δ	E' _{máx} (GPa)	E'' _{máx} (GPa)	ΔE (kJ/mol)
FRPs	Carbon	105.03	100.58	0.45	35.25	8.89	373.88
	Glass	104.13	100.49	0.41	16.73	4.57	408.44
	Sisal	107.43	105.29	0.23	7.88	1.11	445.01
Non-hybrid FMLs	[Al/C ₆ /Al]	109.31	104.80	0.42	19.83	3.53	473.15
	[Al/G ₆ /Al]	98.37	93.58	0.39	13.32	2.02	507.47
	[Al/S ₄ /Al]	107.09	105.65	0.32	14.68	2.86	578.94
Hybrid FMLs	[Al/C ₂ /S ₂ /C ₂ /Al]	106.81	105.40	0.27	7.52	1.22	574.69
	[Al/S ₁ /C ₄ /S ₁ /Al]	107.66	104.89	0.33	8.91	1.53	506.42
	[Al/G ₂ /S ₂ /G ₂ /Al]	96.46	93.51	0.36	12.58	2.20	553.66
	[Al/S ₁ /G ₄ /S ₁ /Al]	101.19	99.55	0.23	5.54	0.77	516.26

Based on Table 10, the T_g values obtained from the Tan δ peak ranged between 95 and 110 °C. This small variation indicates that the glass transition of the system did not undergo significant changes among the different laminate types, demonstrating that neither the presence of aluminum nor the hybridization between synthetic fibers and sisal fiber significantly modified the thermal behavior of the epoxy matrix. Similar results were reported by Benedetto *et al.* [144] who, upon evaluating poly(ether ether ketone) (PEEK) composites reinforced with carbon fiber, observed that the incorporation of different types of steel did not promote significant changes in the thermal stability of the material.

Regarding the activation energy results, the presence of aluminum, combined with the interaction between fibers of distinct natures, increased the energy required for the onset of segmental mobility of the matrix. This behavior indicates that the additional interfaces formed by different materials, particularly in hybrid systems, increased the degree of constraint on the polymer chains, limiting their capacity to rearrange. Furthermore, in laminates containing natural fibers, this trend became even more pronounced, since the intrinsic heterogeneity of sisal, combined with the confinement imposed by the metallic layers, intensifies local stiffness and requires more energy for molecular relaxation.

Regarding stiffness, the carbon fiber-reinforced laminate exhibited the highest E' value (35.25 GPa). This result reflects the high elastic modulus characteristic of carbon fibers and the good interfacial interaction with the epoxy matrix, which favors efficient transfer of the applied stresses. With the incorporation of aluminum into the structure, forming the [Al/C6/Al] laminate, a reduction of approximately 43.8% in E' was observed, decreasing to 19.83 GPa. This behavior can be attributed to the presence of the metallic layers, which alter the stress distribution within the laminate and reduce the direct contribution of the fibrous reinforcement to the overall stiffness of the system.

For the glass fiber-reinforced laminate, an E' of 16.73 GPa was observed, a value lower than that of the carbon laminate but still indicative of good stiffness. This result is also associated with the intrinsic modulus of the glass fiber, which, although lower than that of carbon fiber, remains high compared to natural reinforcements. Moreover, the strong adhesion between the glass fiber and the epoxy matrix facilitates efficient stress transfer, thereby enhancing the mechanical performance of the laminate.

Similarly, when aluminum is incorporated into the [Al/G6/Al] system, the E' value decreased to 13.32 GPa. Although this decrease is less pronounced than that observed for the carbon laminate, it demonstrates that the presence of the metal also influences the overall stiffness of the material. As noted by Xiao *et al.* [145], the incorporation of metallic layers into polymeric composites influences the material's viscoelastic response, modifying the stress distribution between phases and the mechanisms of energy dissipation. As a result, changes are observed in the stiffness and dynamic behavior of the laminate.

For the sisal fiber-reinforced laminate, the E' value was the lowest among the FRPs, at 7.88 GPa. Natural fibers, such as sisal, exhibit lower stiffness than synthetic fibers. Furthermore, the heterogeneity of the fiber/matrix interface results in less efficient load transfer. In addition, factors intrinsic to plant fibers, such as moisture absorption and morphological variability, can introduce structural discontinuities that compromise interfacial adhesion and, consequently, affect the material's viscoelastic response.

When aluminum was incorporated into the [Al/S4/Al] system, behavior opposite to that observed for the synthetic fiber-reinforced laminates was found. The E' value increased by approximately 86.3%, reaching 14.68 GPa. This result suggests that the presence of aluminum acted as an additional reinforcing element, restricting matrix deformation and enhancing the overall stiffness of the system. Furthermore, the metal may have contributed to better compaction and a lower void content, which favored load transfer and reduced

energy losses through internal dissipation. In this case, the aluminum compensated for the low stiffness of the natural fiber and improved the dimensional stability of the material.

When hybridization of synthetic fibers with natural sisal fiber occurs, the mechanical performance of the composite depends not only on the intrinsic properties of the fibers, but also on the layer configuration within the laminate. Among the hybrid composites analyzed, the [Al/G₂/S₂/G₂/Al] laminate exhibited the highest stiffness, with an E' of 12.58 GPa. In this configuration, the glass fibers are positioned in the outer layers, while sisal occupies the central region. This arrangement favors mechanical performance, as the stiffer glass fibers act directly in absorbing the applied stresses, while sisal contributes to damping and energy dissipation within the core of the laminate. Similar results were reported by Xiao *et al.* [145], who found that incorporating higher-modulus fibers into the outer layers of composites increases the storage modulus of the material.

On the other hand, when the fiber stacking sequence was reversed, [Al/S₁/G₄/S₁/Al], a reduction of approximately 55.9% in E' was observed, decreasing to 5.54 GPa. This decrease is associated with the lower stiffness and strength of sisal fibers, which, when placed in the outer layers, become more susceptible to applied stresses, compromising the integrity of the material.

For the hybrid laminates containing carbon fiber, the E' values were relatively close to each other. For the [Al/C₂/S₂/C₂/Al] configuration, the stiffness obtained was 7.52 GPa, while the [Al/S₁/C₄/S₁/Al] configuration exhibited an E' of 8.91 GPa. These results indicate that, despite the lower stiffness observed for [Al/C₂/S₂/C₂/Al], the presence of carbon in the outer layers favored a more efficient interaction with the epoxy matrix during the curing process, reducing local restrictions on molecular mobility and promoting a more uniform formation of the polymer network.

In general, hybridization between synthetic and natural fibers resulted in a significant reduction in stiffness, a direct consequence of the lower elastic modulus of sisal fiber. For the [Al/C₂/S₂/C₂/Al] laminate, for example, a reduction of approximately 78.6% was observed relative to the carbon FRP laminate. A similar trend was reported by Salman [146], who observed a decrease in the storage modulus when combining carbon and jute fibers. According to the same author, this reduction occurs because the presence of natural fibers introduces heterogeneity at the interface and limits the effective contribution of the synthetic fibers, thereby reducing the composite's overall capacity to store energy under dynamic loading.

Regarding the loss modulus (E'') results, a trend similar to that of the storage modulus was observed. The FRP group laminates exhibited the highest E'' values, with the carbon composite reaching 8.89 GPa, with the peak located at approximately 100.58 °C, followed by the glass laminates (4.57 GPa at 100.49 °C) and sisal (1.11 GPa at 105.29 °C). In these materials, composed exclusively of fibers and a polymeric matrix, energy dissipation occurs more markedly because the viscoelastic behavior is strongly influenced by the matrix response and the structural efficiency of the fibers.

Upon incorporation of aluminum and fiber hybridization, a reduction in E'' values were observed. For the non-hybrid FMLs, these values ranged from 2 to 4 GPa, with peaks at 95-110 °C. For the hybrid FMLs, the values were even lower, ranging between 0.5 and 3 GPa. This behavior is related to both the presence of metallic layers and the combination of fibers of distinct natures, which introduce interfacial discontinuities and restrict matrix deformations.

Similarly, the $\tan \delta$ results obtained for the synthetic fiber-reinforced laminates remained within a very close range after the incorporation of aluminum. The carbon FRP exhibited a $\tan \delta$ of 0.45, while the [Al/C₆/Al] laminate recorded 0.42. Similarly, the values observed for the glass fiber-reinforced system were 0.41 for the FRP and 0.39 for the [Al/G₆/Al] laminate. These results show that the addition of the metallic layers did not promote significant changes in the viscoelastic behavior of the materials, keeping their energy dissipation capacity practically unchanged.

In contrast to the behavior observed for synthetic fibers, sisal exhibited an opposite trend. The sisal FRP exhibited a $\tan \delta$ of 0.23, which increased to 0.32 in the [Al/S₄/Al] laminate, indicating that the presence of aluminum modified the fiber/matrix interaction, thereby intensifying damping losses. According to Salman [146], good adhesion between the matrix and the reinforcement tends to limit the mobility of the molecular chains, reducing the damping factor. In natural fibers, however, adhesion to the epoxy matrix is less efficient, which favors greater energy dissipation mechanisms during deformation.

2.5 CONCLUSIONS FROM CHAPTER II

Based on the results obtained, the dynamic-mechanical behavior of the analyzed materials was found to be strongly influenced by both the characteristics of the polymeric matrix and the presence, nature, and distribution of fibers within the laminate. Dynamic mechanical analysis (DMA) proved to be an efficient tool for understanding viscoelastic mechanisms, enabling the simultaneous assessment of stiffness and energy dissipation in the studied systems.

In sisal/epoxy composites, curing conditions played a decisive role in the material's dynamic-mechanical performance. Systems subjected to room-temperature curing followed by post-curing exhibited superior overall performance, characterized by higher storage modulus values and greater thermal stability. Conversely, the use of an accelerator, although it increased T_g , resulted in a reduction in material stiffness. Furthermore, fiber orientation proved to be a critical factor, as 0° -oriented composites exhibited greater load-transfer efficiency and, consequently, superior dynamic-mechanical performance.

In hybrid laminates, viscoelastic behavior was directly related to the structural configuration of the laminate. The presence of synthetic fibers contributed significantly to increased stiffness and structural stability, whereas sisal fibers exerted a greater influence on energy dissipation mechanisms. Additionally, the incorporation of metallic layers into the hybrid laminates altered the redistribution of internal stresses, directly affecting phenomena such as the overshoot observed in E' .

Furthermore, in hybrid FMLs, the position of sisal fibers within the laminate proved to be a determining factor for material behavior. When positioned at the core, the natural fibers were subjected to greater confinement, which promoted the accumulation of internal stresses and led to more abrupt energy releases during heating. In this configuration, the laminates exhibited higher stiffness, directly associated with the presence of synthetic fibers in the outer layers, which bear most of the applied load and restrict system deformation.

In contrast, when sisal fibers were positioned in the outer regions, the synthetic fibers occupied the inner layers of the laminate, reducing their contribution to resistance against applied stresses. In this configuration, a more homogeneous stress redistribution was observed, with a lower incidence of instabilities in the mechanical response, albeit accompanied by a reduction in the system's overall stiffness.

Overall, the results demonstrated that the dynamic-mechanical performance of composites and hybrid laminates is not solely determined by the nature of the constituent

materials, but also depends, in an integrated manner, on the control of processing conditions, the quality of the fiber/matrix interface, and the structural configuration of the system. In this regard, the appropriate combination of matrix, reinforcement, and laminate architecture is essential for achieving materials with superior mechanical properties.

3 FINAL CONCLUSIONS

This dissertation aimed to contribute to the understanding of the residual mechanical and dynamic-mechanical behavior of sustainable composites reinforced with sisal fibers. Although the two chapters addressed different loading conditions and distinct structural systems, both were centered on the use of sisal fiber as reinforcement in polymeric matrices.

Overall, the results obtained indicated that the performance of sisal fiber-reinforced composites was influenced by the processing conditions, the curing state of the polymeric matrix, the quality of the fiber/matrix interface, and the structural configuration of the laminates. In Chapter I, a tendency toward changes in thermal and mechanical properties was observed after creep loading. The results suggested that the residual response of the composites was associated with both the possible progression of residual matrix curing and the quality of the fiber/matrix interface, factors that influenced the performance of the evaluated materials in distinct ways.

In Chapter II, dynamic-mechanical analysis showed that the viscoelastic response of the materials was strongly influenced by curing conditions, fiber orientation, and laminate architecture. In the sisal/epoxy composites, the presence of natural fibers increased the system's stiffness, especially when aligned with the main loading direction. In hybrid fiber-metal laminates, the combination of sisal fibers, synthetic fibers, and metallic layers was found to significantly modify the mechanisms of energy storage and dissipation.

Overall, the results confirm that sisal fibers are a promising alternative for developing sustainable composites. Despite the inherent limitations of lignocellulosic fibers, the results demonstrate that when combined with appropriate processing conditions and suitable structural configurations, these fibers can significantly contribute to the development of materials with mechanical, thermal, and viscoelastic properties compatible with various technological demands.

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