Research Article

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Experimental Investigation of Mechanical Properties of *Myristica fragrans* Seed Filler-reinforced Biopolymer Materials

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ABSTRACT

The research explored the impact of blending *Myristica fragrans* filler with epoxy resin at varying volumes (10%, 15%, 20%, 25%, and 30%) on the mechanical, dynamic mechanical, biodegradability, and thermal properties. These composites were fabricated using compression molding. Results indicated that the composite containing 20% natural filler exhibited superior mechanical properties, storage modulus, and glass transition temperature, along with a reduced tan δ peak. Thermal analysis confirmed enhanced thermal stability due to the addition of natural filler. Biodegradability tests revealed that integrating bio-gum into the composite reduced its weight loss in soil burial experiments.

Keywords: Mechanical properties; Epoxy resin; Composite materials; *Myristica fragrans*; Biodegradability

1. INTRODUCTION

Natural composites, formed by combining natural fibers with synthetic matrices, have gained significant attention in recent years due to their promising properties and environmental benefits. Among these, epoxy resin has emerged as a favored matrix due to its excellent mechanical performance, thermal stability, and adhesive qualities. The integration of natural fillers, such as plant fibers, into epoxy resin not only enhances the composite's mechanical properties but also promotes sustainability by reducing reliance on petroleum-based materials. Natural fillers are biodegradable and renewable, making them an attractive alternative to traditional synthetic reinforcements. This shift towards eco-friendly materials aligns with global efforts to mitigate environmental impact and promote sustainable practices in various industries, including construction, automotive, and packaging. The unique characteristics of natural fibers, including lightweight nature, high specific strength, and good thermal insulation, complement the properties of epoxy resins, resulting in composites that exhibit improved performance across multiple domains. Additionally, the incorporation of natural fillers can enhance the composites' aesthetic appeal and contribute to their overall functionality.This paper aims to explore the advancements in natural composite technology, focusing on the interplay between natural fillers and epoxy resin, and the resulting implications for mechanical performance, thermal stability, and biodegradability. Through comprehensive analysis and experimentation, we aim to provide insights into the potential applications and future directions for these sustainable materials in various sectors.

In the study by (Temesgen *et al.* 2023), mixed acacia and frankincense natural gum-based bioresins were used to develop green composites from enset fibers. These composites show enhanced mechanical strength for lightweight applications, particularly in agro-textiles and geo-textiles, and are cost-effective and easy to produce. Enset fibers exhibit high tensile strength (~500 MPa) and a porous structure with a density of 1.09–1.2 g/cm³, facilitating effective resin absorption. Mechanical testing indicated that tensile strength improved with increasing enset woven fabric up to 30%, achieving a maximum of 4.68 MPa. Beyond this ratio, tensile strength decreased and void content increased, with the optimal ratio being 30:70 enset woven fabric to bioresin. The average tensile strength for enset fabric was 0.53 MPa (warp) and 0.48 MPa (weft), while single-layer composites showed strengths of 2.35 MPa (warp) and 2.07 MPa (weft). Double-layer composites had strengths of 4.67 MPa (warp) and 3.48 MPa (weft). Thermogravimetric Analysis (TGA) revealed two weight loss stages: water loss from 50°C to 250°C and decomposition of bioresin and fibers starting at 300°C. Enset fibers were thermally stable up to 250°C, with a 9% initial water loss. Major degradation occurred at 320°C and 380°C, with raw enset fibers losing 73.33% of their mass between 225°C and 350°C. Enset fabric-reinforced composites showed weight losses of 11% and 80% in the range 330–410°C. Residuals after complete thermal decomposition were 24.67% for raw fibers and 12% for the composite at 800°C, confirming good thermal stability.

Velmurugan *et al.* (2023) focused on developing and investigating a hybrid composite made from nanoscale CuO and ramie fiber reinforced into green epoxy using the hand layup technique. Ramie fiber, with a length of 30 mm, was incorporated at a 20% weight ratio, and nanoscale CuO, synthesized from Urtica dioica leaf, was added in varying amounts (1.5–7.5% wt%). The mechanical, microstructural, and dynamic mechanical properties of the composite were assessed. Results showed that ramie fiber improved the mechanical strength of the green epoxy but reduced its thermal conductivity. The addition of nanoscale CuO up to 3 wt% enhanced its mechanical properties, but higher loadings (above 3 wt%) led to decreased physical performance. The effective distribution and adhesion of nanoparticles in the polymer matrix can restore the mechanical properties diminished by high CuO loadings. Dynamic Mechanical Analysis (DMA) indicated that ramie fiber reduced the damping effect of the green epoxy, while nanoscale CuO increased it. Ramie fiber loading decreased the storage modulus during the glassy stage but increased it during the elastomeric stage. The moisture absorption of the hybrid composite decreased due to the repellent nature of nanoscale CuO and the acidic pretreatment of the fiber and CuO. Overall, the improved thermal and mechanical characteristics of the biocomposite suggest its potential for use in automotive interior components.

Velmurugan *et al.* (2024) tested hybrid epoxy composites reinforced with banana fiber and eggshell powder (ESP). Their study demonstrates significant improvements in mechanical properties using these eco-friendly fillers. They found that combining banana fiber (25 wt%) and ESP (2.5 wt%) resulted in increases in tensile strength (11%), flexural strength (8.9%), and impact strength (12%) compared to pure epoxy resin. The combination of banana fiber and ESP notably enhanced impact resistance.

Balaji *et al.* (2022) experimentally demonstrated that banana 477 fiber (BF) and particle-reinforced (P) epoxy biocomposites show a significant improvement in tensile strength compared to the plain epoxy (E) composite. Among the tested composites, the E/BF composite achieved the highest tensile strength of 36.2 MPa, surpassing the E, E/BP, and E/BFP composites. This improvement is attributed to better fiber-matrix adhesion and fiber orientation in the E/BF composite. Typically, fiber-reinforced composites offer higher tensile strengths than particle-reinforced ones. In contrast, the tensile strength of the E/BP and E/BFP composites is lower due to weak interfacial bonding between the fibers and matrix polymer, which diminishes strength due to fiber and particle inclusion. Thermal stability analysis reveals that all composites had similar stability up to 250°C, with a 2.5% weight loss. From 325°C to 347°C, the composites lost 20% of their weight due to cellulose and hemicellulose degradation, driven by moisture evaporation. Significant weight loss between 348°C and 461°C was due to the decomposition of hemicelluloses, cellulose, and lignin. At 500°C, all samples underwent complete decomposition of combustible components and char formation. The total weight losses after decomposition were 84.45% for E, 79.11% for E/BF, 83.82% for E/BP, and 82.38% for E/BFP, with the E/BF composite showing the least weight loss, indicating superior fiber assimilation and thermal stability.

Navaneethakrishnan and Athijayamani, (2016) produced composite materials using various reinforcements in vinyl-ester resin, including coconut shell powder, sisal fiber, and combinations thereof. They assessed the influence of drilling parameters on thrust force and torque, finding these to be contingent upon manufacturing variables such as feed rate, cutting speed, and tool geometry. Notably, sisal fiber-reinforced composites exhibited higher thrust force and torque compared to other formulations. Ramesha *et al.* (2016) demonstrated that the addition of banyan tree sawdust powder improved the mechanical properties of polypropylene green composites. They also explored the impact of filler material on abrasive water jet machining and reported on the effects of coupling agents and mineral fillers on machining properties (Bootkul *et al.* 2017) utilized teak wood sawdust as a filler in high-density polyethylene, varying the filler content across seven different volume fractions (10% to 70%). Their investigation highlighted that while mechanical properties initially improved with filler content, impact strength decreased notably beyond 30% filler content. Furthermore, they utilized these composite samples in the construction of outdoor decorations for Thai spirit houses. Lette *et al.* (2018) fabricated wood polymer composites using phenolic resin as the matrix and sawdust and rice husk in powder form as reinforcements, maintaining a 60:40 matrix-to-filler ratio. They investigated how exposure to ultraviolet light affected the water stability and mechanical properties of these composites. The results showed that phenolic resin composites reinforced with sawdust exhibited superior mechanical properties compared to those reinforced with rice husk. Sawdust demonstrated better adhesion within the phenolic resin matrix than particulate rice husk filler material. Jaya *et al.* (2018) explored the impact of wood dust powder on the mechanical properties of unsaturated polyester matrices. Their findings indicated that composites containing 6% sawdust powder showed enhanced mechanical performance. They highlighted wood powder's potential as an effective reinforcement and emphasized its feasibility for large-scale composite production. Krishna *et al*. (2018) utilized fly ash and sawdust as filler materials in epoxy resin to develop hybrid composites, varying the composition of fly ash and sawdust. They analyzed the effect of filler loading on hardness, compressive strength, and moisture absorption. Their findings suggested that increasing filler loading beyond a certain threshold decreased these properties due to inadequate wetting of the reinforcements by the epoxy resin. Suthan *et al.* (2019) investigated the mode 1 fracture toughness of epoxy composites reinforced with surface-modified and unmodified sawdust powder, along with woven jute fiber. Various volume fractions of sawdust and jute fiber were incorporated into the epoxy matrix. The experimental findings indicated that composites containing three layers of jute fiber and 20% treated sawdust demonstrated superior mode 1 fracture toughness. Hiremath *et al.* (2018) developed glass fiber-reinforced composites using eggshell powder as a filler material. Composites were fabricated with 5% and 10% eggshell powder alongside 50% glass fiber and the remaining polymer matrix using the hand layup process. Their study of mechanical and physical properties revealed that stiffness increased notably with 10% eggshell powder incorporation. Ganesan *et al.* (2018) produced polyester matrix composites reinforced with jute fiber, employing both NaOH-treated and untreated jute fabric. They investigated the influence of nano clay and eggshell powder additives. Results indicated that the composite formulation containing 1.5% nano clay and 1.5% eggshell powder exhibited superior mechanical properties compared to other compositions. Chemical treatment of the reinforcement enhanced interfacial adhesion and overall mechanical performance.

2. EXPERIMENTAL DETAILS AND TESTING METHODS

2.1 Materials

In this research, *Myristica fragrans* filler was incorporated into both natural epoxy resin and LY 556 epoxy resin. The density of the *Myristica fragrans* filler was measured at 1.06 g/cm³. After ball milling for 4 hours, the filler was ground to achieve a particle size ranging from 5 to 10 microns. Natural resin was subsequently added to the epoxy matrix in varying proportions (10%, 15%, 20%, 25%, and 30%) and thoroughly mixed for 45 minutes using a mechanical blender to ensure even distribution. Compression molding was employed to create laminates of the *Myristica fragrans* filler with epoxy hybrid polymer material. After curing, the laminates were cut in accordance with ASTM (American Society for Testing and Materials) standards for a range of tests.

2.2 Mechanical Properties

Tensile and flexural testing were carried out utilizing an Instron Universal Testing Machine. The tensile test followed ASTM D-638 guidelines at a speed of 2 mm/min. Flexural strength was evaluated using a three-point bending test in accordance with ASTM D-790 guidelines. The Izod method, as specified in ASTM D-256, was used to assess impact strength. Each test was carried out on five samples, and the average result was calculated.

2.3 Dynamic Mechanical Analysis

The dynamic properties of a *Myristica fragrans* filler blended epoxy hybrid polymer material was examined using the SEIKODMAI-DMSC 6100 under conditions of heat and dynamic loading. Dynamic mechanical analysis tests were conducted in a nitrogen environment, employing the tensile mode. The tests covered temperatures ranging from 30 to 180℃, with temperature increments of 5℃/min and a frequency of 10 Hz.

2.4 Thermogravimetric Analysis

The composite specimen's thermal stability was assessed using a TG/DTA 6200 SEIKO TGA analyzer. The tests were carried out at a heating rate of 20℃/min in the temperature range of 0 to 800℃. To avoid oxidation, the material was heated in a nitrogen atmosphere.

2.5 Biodegradability Test

The traditional and widely accepted method for assessing degradation is soil burial. Soil, being a complex and dynamic ecosystem due to diverse waste disposal conditions, hosts a multitude of organisms that actively contribute to degradation processes. To investigate biodegradability, samples were initially weighed and then bured in soil. At intervals of 5, 10, 20, 30, 40, 50, 60, 70, 80, 90, 100, 110, and 120 days, the buried samples were excavated. After the samples were removed from the soil, they were washed with water and weighed to determine the amount of weight loss.

2.6 Fractography Study

The dispersion of the filler material within the polymer matrix was assessed using a scanning electron microscope. A Hitachi S 3400 N scanning electron microscope was employed for this purpose. To prevent charging, the composite samples were coated with a thin layer of gold via sputter coating.

3. RESULTS AND DISCUSSION

This work proposes integrating *Myristica fragrans* filler with epoxy resin to enhance the properties of hybrid polymer materials. A thorough examination was conducted to compare these results with those of epoxy resin alone.

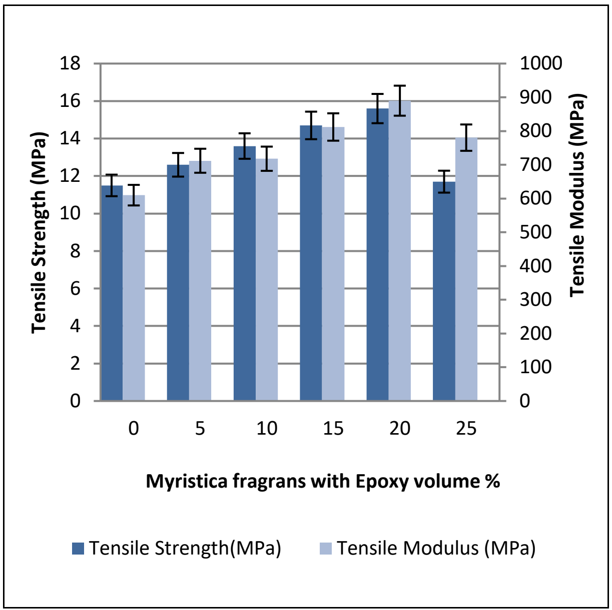

Fig. 1: Tensile Property of *Myristica fragrans* with epoxy blender hybrid material

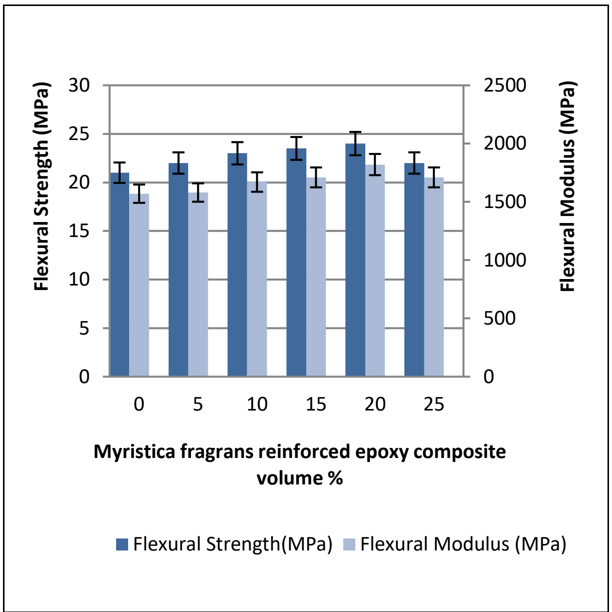

Fig. 2: Flexural properties of *Myristica fragrans* blended epoxy with epoxy blender hybrid material

Fig. 1-3 illustrate the mechanical properties of hybrid polymer composites consisting of *Myristica fragrans* filler blended with epoxy resin. Fig. 1 details the tensile characteristics of the composites as a function of natural resin added to epoxy resin. Pure epoxy resin exhibits a tensile strength of 17.5 MPa and a modulus of 1200 MPa. With the addition of natural resin at 10, 15, 20, 25, 30, and 35 volume percent, the tensile strength increases to 18.644, 19.845, 21.46, 23.56, 25.17, and 24.13 MPa, respectively. Similarly, the tensile modulus increases from 1370, 1410, 1444, 1612, 1712, to 1590 MPa at 30% composite. The strength and modulus of *Myristica fragrans* filler blended with epoxy bio-resin improve by 12.26% and 31.97%, respectively, with a 30-volume percent addition of the filler. Moreover, increasing the addition to 35 volume percent decreases both strength and modulus from 25.17 MPa to 24.13 MPa. Fig. 4 and 5 display micrographs of hybrid polymer composites with 30% and 35% natural resin added.

Fig. 2 illustrates the flexural characteristics of the composite. The flexural strength and modulus of the epoxy resin are 34 and 1700 MPa, respectively. With the addition of *Myristica fragrans* filler blended epoxy natural resin at 10, 15, 20, 25, 30, and 35 volume percent, the flexural strength increases to 34.56, 35.74, 36.26, 37.45, 41.72, and 39.92 MPa, respectively. Similarly, the flexural modulus increases to 2010, 2600, 2850, 3100, 3570, and 2419 MPa at these volume percentages. The composite with 30% *Myristica fragrans* filler blended epoxy shows a 15.37% increase in flexural strength and a 22.70% increase in flexural modulus. The flexural properties of *Myristica fragrans* mixed with epoxy hybrid polymer materials exhibit improvements up to 30% volume percent, similar to tensile properties.

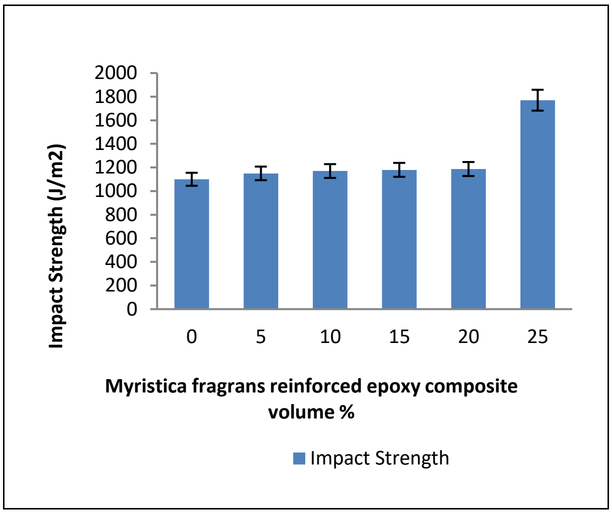

Fig. 3: Impact properties of *Myristica fragrans* blended epoxy with epoxy blender hybrid material

Fig. 3 displays the impact strength of the *Myristica fragrans* filler blended epoxy hybrid polymer material. The impact strength of the epoxy resin is 2 J/m², while the impact strengths of the hybrid polymer materials with 10, 15, 20, 25, 30, and 35 volume percent are 2469, 2557, 2648, 2687, 2718, and 2671 J/m², respectively. The hybrid polymer composite with 30-volume percent *Myristica fragrans* filler blended epoxy exhibits the highest impact strength of 2718 J/m². The lower impact strength of epoxy resin is attributed to its brittleness, whereas the *Myristica fragrans* filler blended epoxy shows enhanced adhesion with the epoxy resin, leading to improved ductility. Consequently, the composite exhibits an 18.4% increase in impact strength.SEM image (Fig. 4) illustrates hybrid polymer materials with 25 volume percent natural resin added, showing even distribution of the natural resin throughout. This uniform dispersion enhances the load-carrying capacity of the hybrid polymer composites.

Fig. 5 reveal agglomeration due to the higher volume percentage of natural resin. This agglomeration prevents uniform distribution of the resin, leading to degradation in mechanical properties.

3.1 Dynamic Mechanical Analysis

Dynamic mechanical analysis (DMA) is utilized to evaluate the viscoelastic behavior of polymers by assessing their damping characteristics under cyclic loading. It quantifies the energy dissipation within the polymer material. This study investigates the influence of *Myristica fragrans* filler blended epoxy natural resin volume percent on the storage modulus and damping factor of hybrid polymer materials. Additionally, the study examines the impact of frequency variation on the storage modulus of these hybrid polymer materials.

3.1.1 Storage Modulus

Fig. 6 illustrates how temperature and loading affect the storage modulus of a hybrid polymer composite incorporating *Myristica fragrans* filler blended with epoxy natural resin, tested at a frequency of 10 Hz. The findings suggest that adding *Myristica fragrans* blended epoxy natural resin enhances the composite's capacity to absorb energy. This enhancement is attributed to the integration of natural materials into the epoxy matrix, thereby improving the composite's energy storage capabilities.

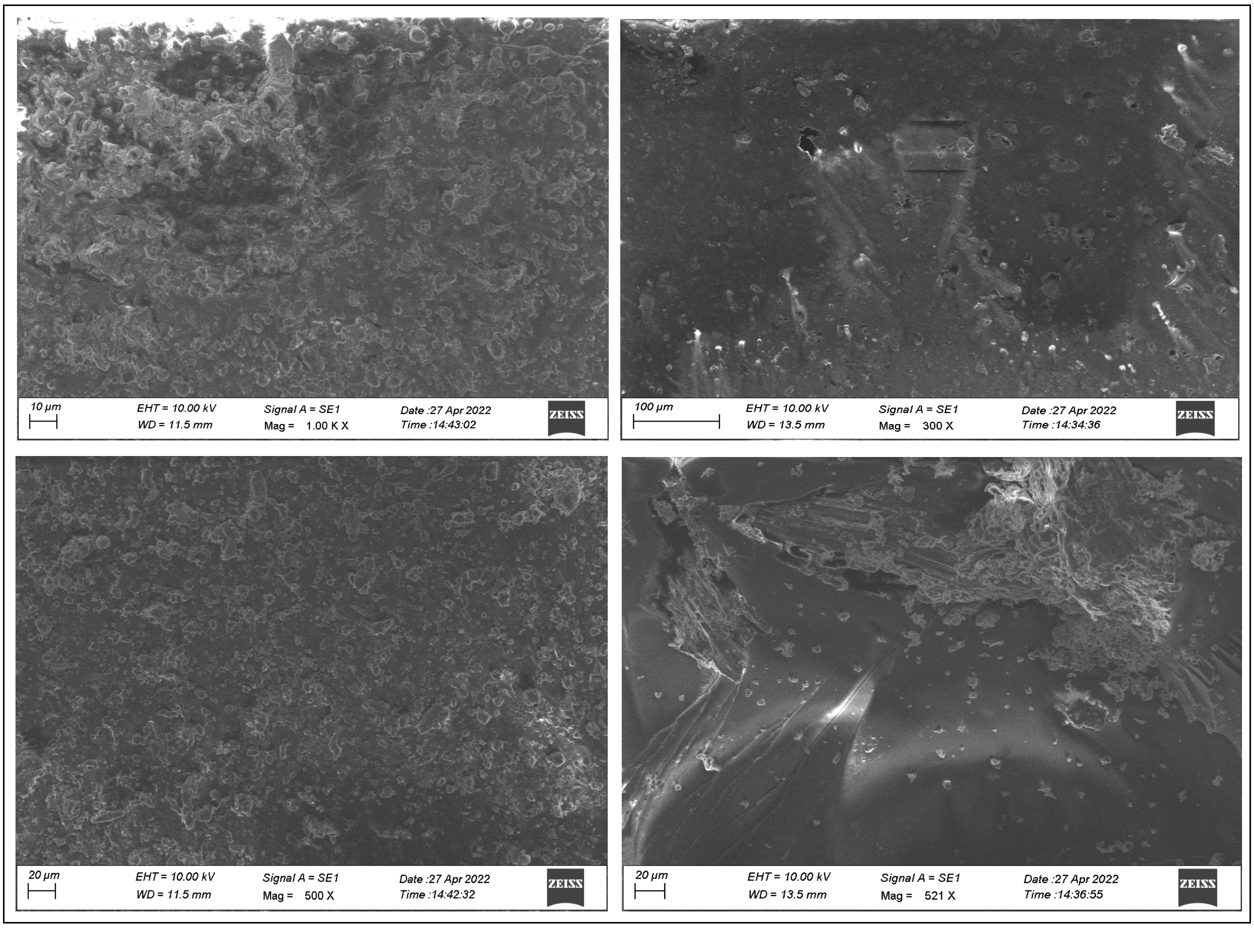

Fig. 4: SEM images of hybrid polymer materials with 25 volume percent natural resin added

Fig. 6 indicates that pure epoxy resin exhibits a lower storage modulus, implying higher stiffness. The glass transition temperature (T_g) of the composite increases from 70 to 110°C, attributed to increased molecular mobility within the polymer chain.

Researchers suggest that incorporating *Myristica fragrans* filler blended epoxy natural resin into the hybrid polymer material enhances stiffness. Another significant finding from Fig. 6 is the utilization of *Myristica fragrans* mixed epoxy natural resin, which enhances the storage modulus in both the glassy and rubbery regions. Studies have demonstrated that shorter exposure times (high frequency) result in higher storage modulus values, while longer exposure times (low frequency) result in lower storage modulus values. This phenomenon is attributed to molecular rearrangement within the material to alleviate localized stresses.

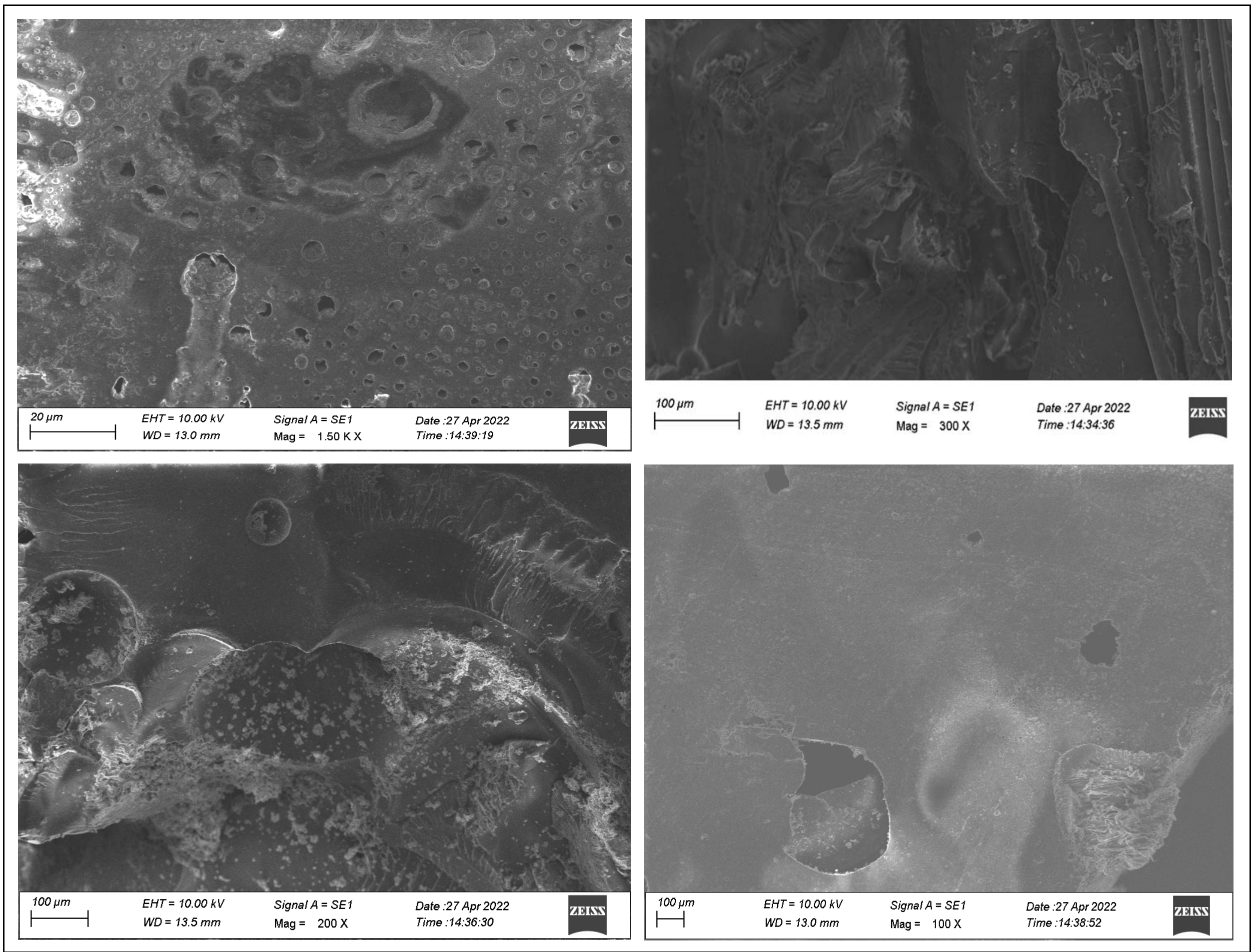

Fig. 5: SEM images of hybrid polymer material with 30 volume percent natural resin

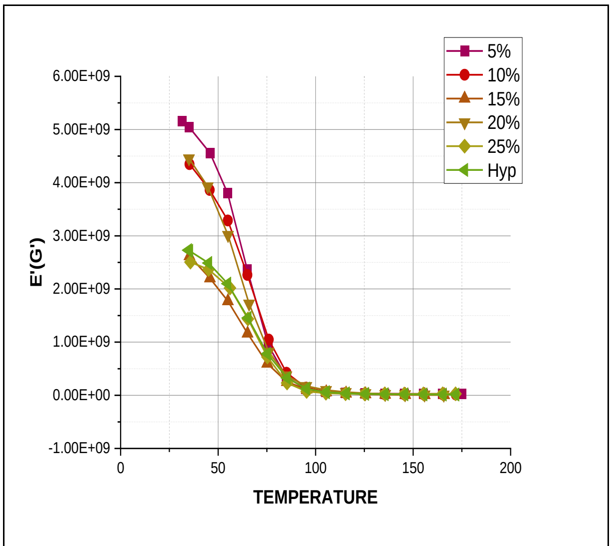

Fig. 6: Storage modulus of *Myristica fragrans* blended epoxy with Epoxy Blender Hybrid at 10 Hz Frequency

3.1.2 Damping Factor (Tan δ)

The damping factor, which represents the ratio of the material's loss modulus to its storage modulus, indicates the energy dissipation and molecular mobility within the polymer chain under loading. Fig. 7 and 8 illustrate the damping factor of the hybrid material incorporating *Myristica fragrans* blended epoxy natural resin. The findings indicate that adding *Myristica fragrans* blended epoxy natural resin to the polymer mixture up to 30% enhances the damping factor. This suggests that polymer composites with lower percentages of *Myristica fragrans* filler blended with epoxy natural resin exhibit greater interaction between synthetic and natural resins, leading to increased energy dissipation of natural materialsHowever, as depicted in Fig. 7 and 8, higher volumes of natural resin (exceeding 30% volume percent) reduce energy dissipation. Compared to the hybrid polymer material with added *Myristica fragrans* filler blended epoxy natural resin, the peak height of Tan δ (damping factor) is lower for neat epoxy resin. This indicates that higher loading of natural resin increases the rigidity of the composite material.

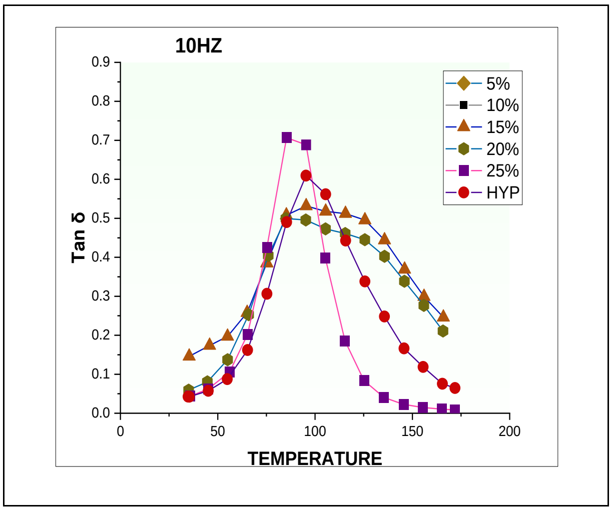

Fig. 7: Tan δ of *Myristica fragrans* blended epoxy with epoxy blender hybrid material at 10 Hz frequency

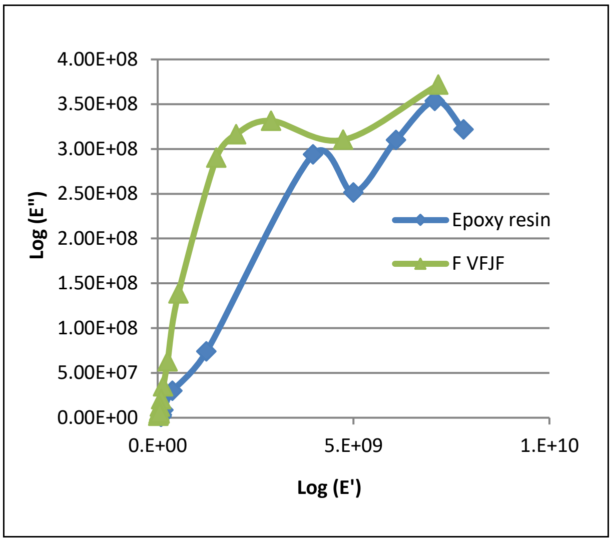

Fig. 8: Cole- Cole Plot *Myristica fragrans* blended epoxy with epoxy blender hybrid material

3.1.3 Cole-Cole Plot

The Cole–Cole plot is a useful tool for analyzing the linear viscoelastic behavior of polymer composites at their glass transition temperature, plotted typically as loss modulus (E″) versus storage modulus (E′) at 10 Hz frequencies. Incorporating natural *Myristica fragrans* filler blended epoxy resin induces structural changes in the cross-linked polymer, as depicted in this plot. The nature of the curve on the Cole–Cole plot indicates whether the material is homogeneous or heterogeneous.

Fig. 8 demonstrates that the hybrid polymer material exhibits homogeneous characteristics. Specifically, mixing *Myristica fragrans* filler blended epoxy natural resin with 30% volume results in a homogeneous substance. However, adding *Myristica fragrans* filler blended epoxy natural resin at 30% volume causes the material's behavior to transition from homogeneity to heterogeneity. In the Cole–Cole plot, a semicircular curve is observed at 30% volume, and any deviation from this semicircular nature suggests increasing heterogeneity in the composite.

3.1.4 Thermogravimetric Analysis

Thermogravimetric analysis (TGA) curves comparing neat epoxy resin with the hybrid polymer material incorporating *Myristica fragrans* blended epoxy resin. The results indicate that the hybrid polymer material exhibits greater thermal stability compared to pure epoxy resin. Incorporating *Myristica fragrans* blended epoxy resin into the epoxy matrix does not significantly affect thermal stability, although there is a slight enhancement in TGA values due to the components of *Myristica fragrans* blended epoxy natural resin.

The initial stage of weight loss in the TGA curves is attributed to water evaporation from the sample. Importantly, the addition of *Myristica fragrans* blended epoxy natural resin to the epoxy matrix enhances the thermal stability of the hybrid polymer materials

In Fig. 9, the thermal stability of epoxy resin is illustrated up to 491°C, where it rapidly decomposes, leaving only 0.3% residue. In contrast, *Myristica fragrans* blended epoxy resin shows stability up to 490°C with a residual content of 9%, demonstrating its improved thermal stability up to 30% volume percent.

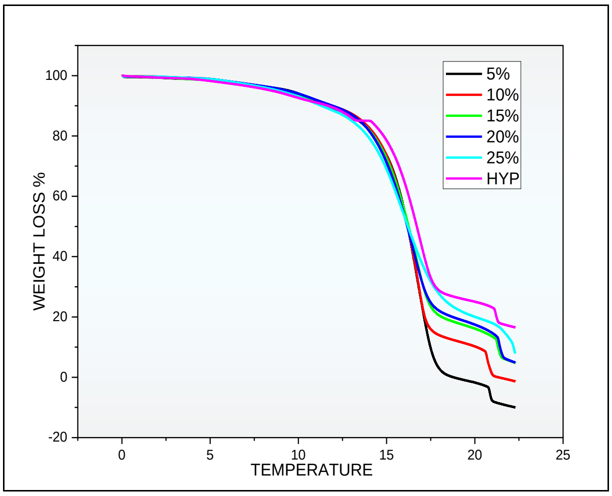

Fig. 9: Thermogravimetric analysis of *Myristica fragrans* with epoxy resin

3.1.5 Biodegradability Test

Fig. 10 illustrates the biodegradability of *Myristica fragrans* blended epoxy hybrid polymer materials. Initially, the sample's weight was recorded before burying it in damp soil. Over 120 days, periodic measurements showed a 5.2% reduction in its overall weight. Initially, the sample absorbed moisture, causing an increase in weight, followed by a gradual decrease due to microbial and environmental interactions.

The weight loss observed over time is attributed to the activity of micro- and macro-organisms interacting with the composite. Comparative analysis with other natural fiber and filler-reinforced polymer composites indicates that *Myristica fragrans*-blended epoxy hybrid material exhibits enhanced biodegradability. Consequently, utilizing *Myristica fragrans* filler blended epoxy hybrid materials contribute to the development of more environmentally friendly polymers.

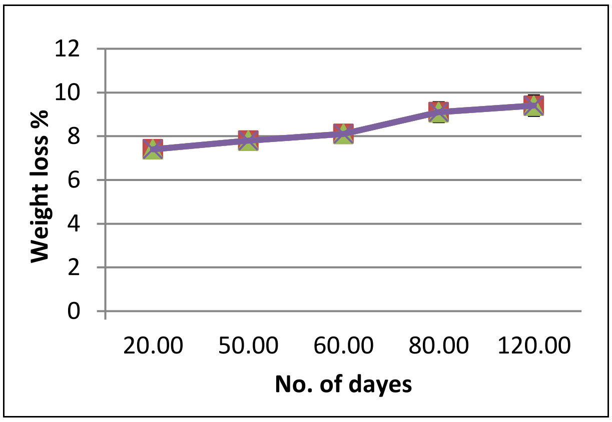

Fig. 10: Biodegradability of *Myristica fragrans* blended epoxy hybrid polymer materials

4. CONCLUSION

The study investigated the impact of varying volumes of *Myristica fragrans* filler mixed with epoxy on mechanical, thermal, biodegradability, and dynamic properties, including storage modulus and damping factor. Mechanical testing revealed that incorporating 30% filler into the *Myristica fragrans* blended epoxy matrix enhanced composite properties such as tensile, flexural, and impact strength. Similarly, dynamic mechanical analysis showed an increase in storage modulus and glass transition temperature with the 30% addition, indicating improved stiffness between the matrix and filler material.

However, SEM images indicated that filler inclusion led to uneven distribution within the *Myristica fragrans* blended epoxy system, potentially reducing material homogeneity. Furthermore, biodegradability testing demonstrated a significant weight loss of 5.2% over 120 days, highlighting the composite's biodegradable nature.

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CONFLICT OF INTEREST

The authors declared no conflict of interest in this manuscript regarding publication.

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