Research Article

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Experimental investigation of *Camellia sinensis* Dust Filler on the *Vachellia nilotica*-Blended Hybrid Epoxy Composites: A Comprehensive Analysis of Mechanical, Viscoelastic, and Dielectric Behavior

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ABSTRACT

This research work presents the influence of *Camellia sinensis* dust powder on the mechanical, dynamic, and dielectric properties of the hybrid epoxy composites containing *Vachellia nilotica*. The hand layup method was used to make the composite specimens at different volume fractions of the *Camellia sinensis* dust used as a particle filler (3, 6, 9, 12, and 15 v/v%). The *Vachellia nilotica* content was kept at 15 v/v%. The mechanical, viscoelastic, and dielectric strengths of hybrid composites were investigated experimentally. The results indicate that the composite with 9 v/v% *Camellia sinensis* dust has superior mechanical and dielectric properties when compared to other volume percentages. The epoxy composite with the highest glass transition temperature and storage modulus also contained 9 v/v% filler. The inclusion of *Camellia sinensis* dust in the *Vachellia nilotica* incorporated hybrid exposed had improved the thermal stability of the composite, as revealed by thermogravimetric analysis. Furthermore, the dielectric test demonstrated that the addition of *Camellia sinensis* dust filler improved the dielectric strength of the composite. The structural morphology of the composite's tensile fracture was investigated using scanning electron microscopy equipment to study the interaction between the epoxy matrix and the fillers in the hybrid composites.

Keywords: Epoxy composite; *Camellia sinensis*; *Vachellia nilotica* resin; Thermogravimetric analysis; Dielectric properties.

1. INTRODUCTION

Natural fiber composites play a vital role in the automotive, packaging and furniture industries. Several researchers (Vijay *et al.* 2020; Khan *et al.* 2022; Borah *et al.* 2023) have studied natural fiber and natural filler incorporated polymer composites. These composites are being explored due to several advantages such as low weight, low cost, weight specific strength, ecofriendliness, ease of availability and recyclability (Vinod Kumar *et al.* 2017; Yesuraj *et al.* 2021). The rise in industry and urbanization in recent years has contributed to a greater awareness of environmental issues, particularly those pertaining to the handling of industrial and agricultural waste (Palanikumar *et al.* 2018; Gokulkumar *et al.* 2021; Debnath *et al.* 2021). A significant amount of waste tea leaves are produced continuously. Tea dust is frequently dumped in landfills or incinerators, which degrades the environment and spoils precious resources. Vijay *et al.* (2020), Prabhu *et al.* (2022) and Kartal *et al.* (2024) studied epoxy composite reinforcements with *Sansevieria ehrenbergii* (SGF), waste tea leaf (WTLF), and glass fiber. It was reported that composite material with 25% SGF and 5% WTLF has a maximum tensile strength of 79.32 MPa, 26.2% higher than one with 5% SGF and 25% WTLF (62.84 MPa). Superior fiber-matrix adhesion increased composite flexural strength by 28.67%. Compared to 5% SGF, 25% SGF increased impact strength by 77.4%. Interlaminar shear strength was 9.6 MPa for the 25% SGF composite due to strong interfacial bonding. Due to its porous structure, the 25% WTLF composite achieved the highest acoustic absorption coefficient (AAC) of 0.59. Brewed tea waste (BTW) was used to reinforce jute polyester composites by (Aftab et al. 2024). The investigation found that 6 wt% filler produced 25.45 MPa tensile strength, 37.56 MPa flexural strength, and 1463 MPa tensile modulus. Filler agglomeration reduced mechanical characteristics beyond 6 wt%. At 15 wt%, hydrophilic fillers increased bulk density, water absorption, and soil deterioration. Scanning Electron Microscope (SEM) showed strong interfacial solid bonding at 6 wt% filler content, resulting in excellent characteristics. Thermal characteristics showed that 15 wt% filler increased stability. This study suggests that black tea waste could improve composite performance as an eco-friendly filler. The flexural performance of epoxy matrix composites was studied by Anandha Moorthy *et*

al. (2021). Ultrafine solid particles (UCS) and chemically modified and non-specifically modified banana fiber strengthened the composites. The NaOH alkaline treatment improved interfacial bonding between epoxy matrix and banana fiber. The composite containing 65% epoxy, 33% modified banana fiber, and 2% UCS exhibited a maximum flexural modulus of 5311.81 MPa and a maximum flexural strength of 70.50 MPa. By improving fiber-matrix adhesion and chemically removing contaminants, modified banana fiber composites had 20% better flexural performance than unmodified composites. Flexural characteristics improved with increasing distribution of UCS particles, showing that chemical modification and fillers can enhance composite performance. Dutta *et al.* (2019) synthesized waterborne polyester nanocomposites using cellulose nanofibers derived from discarded green tea leaves. Continuous nanofibers (0.25, 0.5, and 1 wt%) were used to make nanocomposites. Glycerol-incorporated epoxy and fatty acid-incorporated poly (amido amine) hardeners were cross-linked. The tensile strength and mechanical characteristics of the nanocomposite improved. Risnasari *et al.* (2019) examined the physical, mechanical, and formaldehyde emissions of particleboard, made from residual tea leaves (*Camellia sinensis*), Meranti wood (*Shorea sp*), and UF glue. The highest recorded value of 36101.48 kgf/cm^2 was reached using a 25:75 tea leaf to Meranti wood ratio and 9% hardener concentration. Higher tea leaf content lowers the modulus of rupture due to lower cellulose and hemicellulose. Researchers achieved a maximum internal bonding strength of 1.12 kg/cm^2 utilizing 100% tea leaf waste and 9% hardener. This investigation found that tea leaf waste boards had the lowest formaldehyde emissions at 0.95 mg/L. Phenolic chemicals in tea leaf react with formaldehyde to form this.

Khodadad Hatkeposhti *et al.* (2024) studied the effect of adding leftover black tea in polylactic acid (PLA) biocomposites to improve mechanical characteristics and sustainability. Black tea waste powder and PLA were mixed at 3 and 5 wt% using a twin-screw extruder. In the impact test, the biocomposite with 3% tea waste exhibited the highest fracture energy of 2.5 kJ/m^2 and a 34% increase in tensile strength of 67 MPa, compared to pure PLA's 50 MPa. The dispersion of tea particles enhanced transverse stress transfer within the matrix. Kushwanth Theja *et al.* (2023) tested the mechanical performance of hand-laid epoxy composites, which improved with the use of tea biofill dust (TD). The composite material with 50% TD exhibited a maximum tensile strength of 38.2 MPa and a modulus of 1700 MPa, outperforming lesser filler levels. At 40% TD, the peak impact strength was 22.8 kJ/m^2, while at 50% TD, the maximum flexural strength was 52.6 MPa. The analysis found that a 50 vol.% TD epoxy balanced strength and durability, making it appropriate for lightweight engineering applications.

Juszkiewicz *et al.* (2024) investigated the biofiller effects of powdered tea waste (GT) in natural rubber (NR) composites. At 15.4 MPa, 10 phr GT has 40% better maximum tensile strength than unfilled NR. This surge is due to matrix-GT interaction and increased dispersion. Agglomeration reduced tensile strength at higher GT concentration. Interfacial layers increased brittleness and crosslink density, reducing tensile strength but increasing filler dispersion. Green tea polyphenols may have inhibited thermo-oxidative aging. The first breakdown temperature dropped 32–52 °C due to GT concentration, affecting thermal stability.

Girimurugan *et al.* (2021) and Gokulkumar *et al.* (2021) showed that *Camellia sinensis* dust particles can be used as effective particulate fillers in refining the mechanical and acoustic properties of polymer composite materials. Chemical treatment improved the surface bonding between the natural filler and the polymer resin. NaOH was used as the chemical agent. This investigation showed that treated fibers and appropriate tea particle content improved composite water absorption resistance. Based on the reported literature, the use of tea dust as a potential biofiller in polymer based composites has been explored. However, there had been very less work on the use of tea dust filler in the hybrid matrix containing epoxy resin and natural resin. In the current work, the influence of *Camellia sinensis* dust filler on the mechanical, Dynamic Mechanical Analysis, dielectric and Thermogravimetric (TGA) properties of epoxy hybrid composites was investigated. Scanning electron microscope (SEM) analysis was carried out to analyze the fracture morphology of failed specimen.

2. MATERIALS AND METHODOLOGY

2.1 Materials

This experiment used *Vachellia nilotica* powder as bio-resin and epoxy resin (Araldite LY 556) with hardener (HY951) as matrix materials. Epoxy was procured from Sakthi Fibers in Chennai. The epoxy and hardener were mixed in a 10:1 weight ratio as per the specifications for optimal curing and mechanical properties. The *Camellia sinensis* dust filler, purchased from Sakthi Fibers in Chennai, has particles averaging 200–500 µm in size. *Camellia sinensis* was chosen as a filler because of its abundant availability, better mechanical properties and low cost, which increase epoxy matrix biodegradability without compromising strength. Powdered *Vachellia nilotica* (Babul) with a particle size of approximately 50–100 µm was procured from Vimal Enterprises in Kanchipuram, India. *Vachellia nilotica*, a biodegradable resin, is known to improve mechanical properties of composite. This study examined the synergistic effects of combining *Vachellia nilotica* and *Camellia sinensis* dust fillers to improve reinforced material performance. The handle up method was used to manufacture the composite samples. The vol. % of the particulate filler was kept at 3, 6, 9, 12, and 15

v/v%. The *Vachellia nilotica* content was kept at 15 v/v% and the remaining was epoxy resin. Following that, the formulation was vacuum degassed to remove any trapped air bubbles that could affect composite properties. Next, the resin mixture was carefully poured into molds and cured at room temperature for 24 hours. A two-hour post-cure at 80 ℃ ensured complete polymerization and improved mechanical properties. The test samples were cut from the composite plate according to American Society for Testing and Materials (ASTM) norms.

2.2 Testing Methods

Instron 5567 universal testing equipment was used to measure the modulus and strength of the polymer composite material according to ASTM D638. The maximum crosshead velocity was kept at 2 mm/min throughout testing. Five specimens were tested to ensure statistical validity, and their average values were carefully recorded and presented. Flexural strength and modulus were tested using the Instron machine with a three-point bending fixture per ASTM D790. Testing uses a 10 mm/min crosshead displacement rate. This method evaluated the composite's mechanical deformation resistance under transverse load. A Ceast Resil Impactor was used to assess Charpy impact strength according to ASTM D256. This experiment used the average impact energy from five specimens to test the material's resistance to sudden force or impact. Dynamic mechanical analysis which is generally used to assess the viscoelastic properties of the polymer composite materials was used in this experiment. The storage modulus (E'), loss modulus (E"), and damping factor (tan δ) were assessed using a SEIKODMAI-DMSC 6100 analyzer. The experimental testing followed ASTM E1640 using a tensile mode at 1 Hz and 5 ℃/min heating. A CEAST 6135 breakdown potential analyzer assessed the dielectric breakdown voltage according to ASTM D149 standards. Circular specimens 80 mm in diameter and 3 mm thick were exposed to 100 V per minute up to breakage.

3. RESULTS AND DISCUSSION

3.1 Mechanical Strength

The tensile, flexural, and impact strengths of hybrid epoxy composites containing varying amounts of *Camellia sinensis* dust filler are displayed in Fig. 1–3. The variation of tensile strengths and the composites' modulus are shown in Fig. 1. The tensile strength of the composite material shows a slight improvement when the amount of *Camellia sinensis* dust filler increases up to 9 v/v% as shown in Fig. 1. The uniform distribution of fillers inside the epoxy matrix and the strong interfacial bonding, which allow for an efficient transfer of load from the matrix to the fillers, are responsible for the observed increase. Conversely, the tensile strength decreased when the filler concentration raised above 9 v/v%.

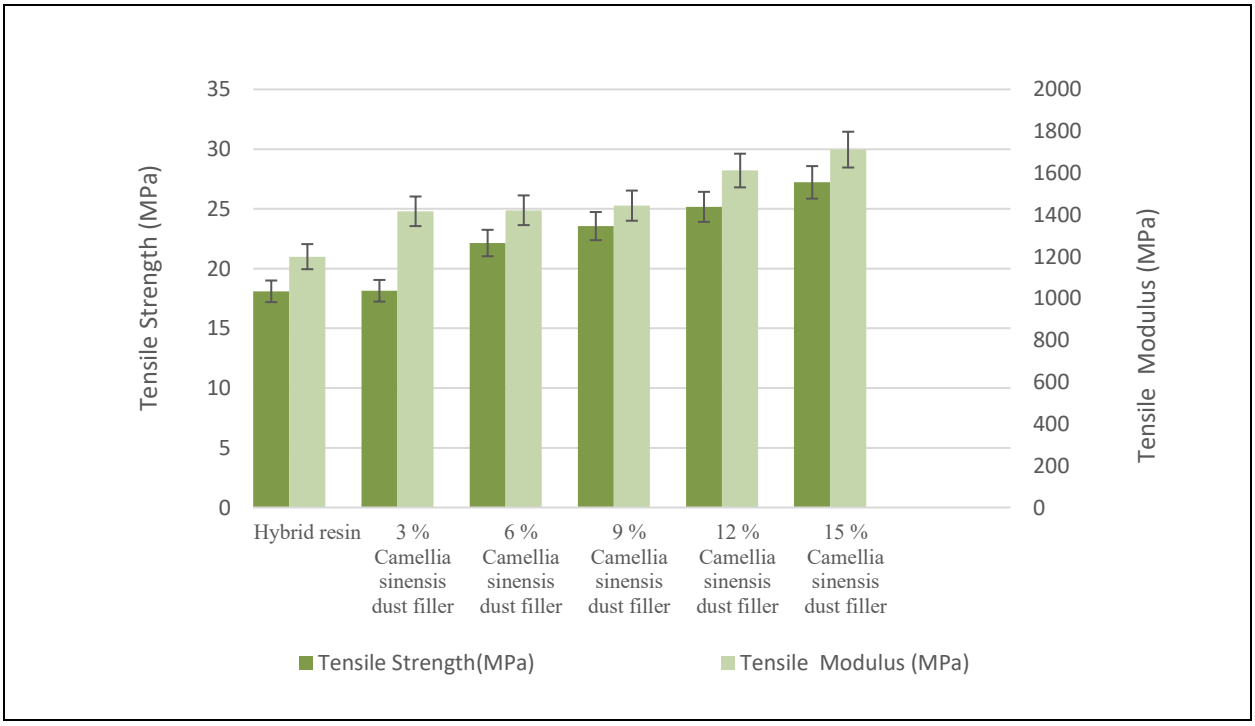


Fig. 1: Tensile strength of *Camellia sinensis* dust filler - *Vachellia nilotica* /epoxy matrix composite

The start of filler agglomeration, which resulted in poor adhesion between the filler and the matrix and the formation of microvoids, is most likely responsible for the reduction. Because of the stress concentration caused by these defects, the composite material's total tensile strength is decreased. The tensile modulus exhibits a discernible pattern, with a steady increase up to a filler content of 9 vol.% and a more notable decline at higher filler volumes. The tensile modulus follows the following pattern. The observed drop in modulus beyond 9 vol.% may result from increased occurrences of weaker interfacial areas between the matrix and the fillers.

The composites' flexural properties followed a similar pattern as that of the tensile characteristics, as illustrated in Fig. 2. A relationship is established between the composite materials' modulus and flexural properties. Adding *Camellia sinensis* dust filler at up to 9 v/v% significantly increases the flexural strength, resulting in an increased capacity to tolerate bending pressures. The uniform dispersion of the particulate fillers, especially those with strong adhesion in the outer layers of the composite material, is responsible for the observed improvement. This alignment increases the composite's flexural strength by efficiently distributing the stress brought on by the application. On the other hand, as the filler content goes above 9 v/v%, the flexural strength decreases. These elements include filler agglomeration and inadequate bonding, both of which compromise the composite's structural integrity under bending forces.

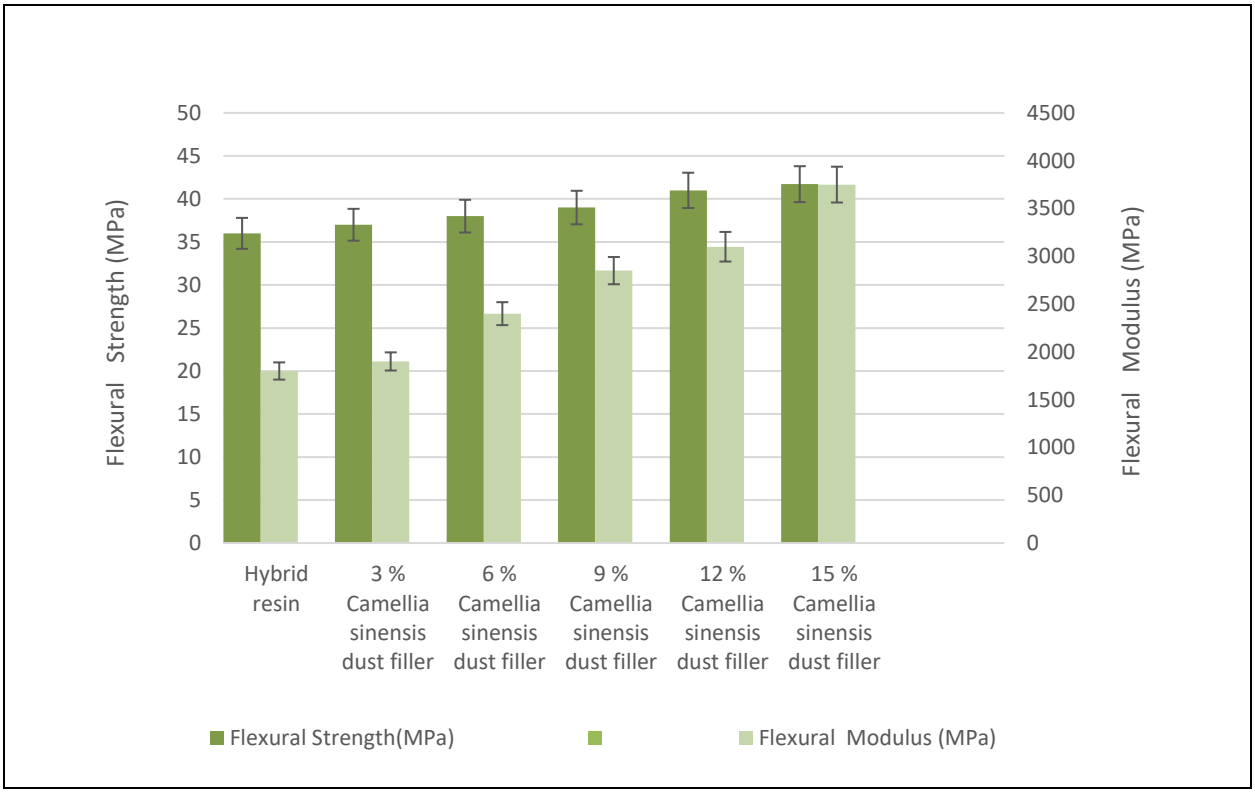


Fig. 2: Flexural strength of *Camellia sinensis* dust filler and *Vachellia nilotica*/epoxy matrix composite

Up to a volume percentage of 9 v/v%, Fig. 3 shows a significant improvement in impact strength when *Camellia sinensis* dust filler is added. The fillers' ability to absorb the energy is mostly responsible for the composite's increased impact strength. Fillers can help bridge and deflect fractures when used in the correct ratios. The effectiveness of these processes leads to a more efficient dispersion of impact energy, which in turn improves the composite's toughness. Conversely, filler agglomeration and the ensuing weak interfacial bonding cause the impact strength to drop when the filler concentration rises above 9 v/v%. The deterioration of the material's tensile and flexural characteristics is analogous to this process.

3.2. Fractography Observation of *Camellia sinensis* Dust Filler Dispersed *Vachellia nilotica* /Epoxy Composites

The SEM analysis of the composites' fracture surfaces provides insights into the failure mechanisms that occur under tensile loading. The micrographs in Fig. 4 (a)–(d) offer significant insights into the interaction between the filler and the matrix at different filler loadings. Fig. 4 (a) shows the tensile fractured sample

with 3 v/v% of the *Camellia sinensis* dust filler, which appears to be uniformly dispersed throughout the epoxy matrix. Based on the SEM image, which shows very little filler clumping.

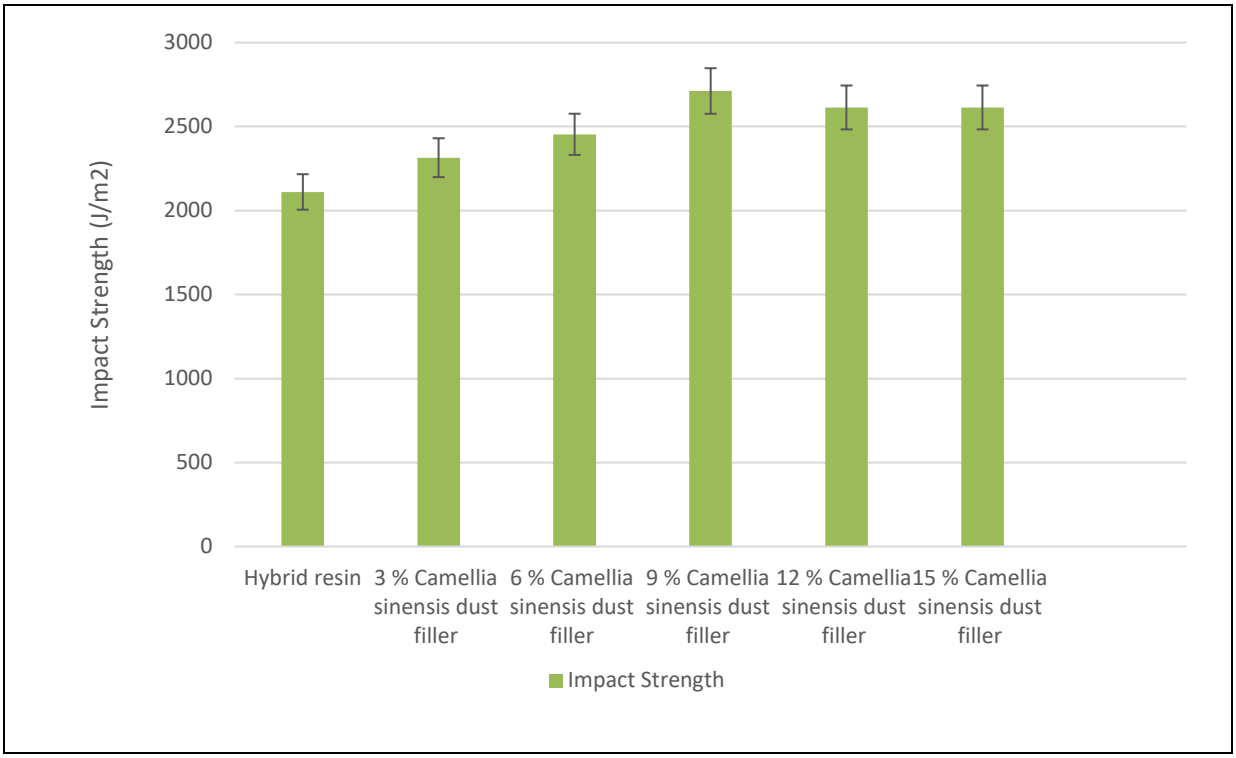


Fig. 3: Flexural strength of *Camellia sinensis* dust filler and *Vachellia nilotica*/epoxy matrix composite

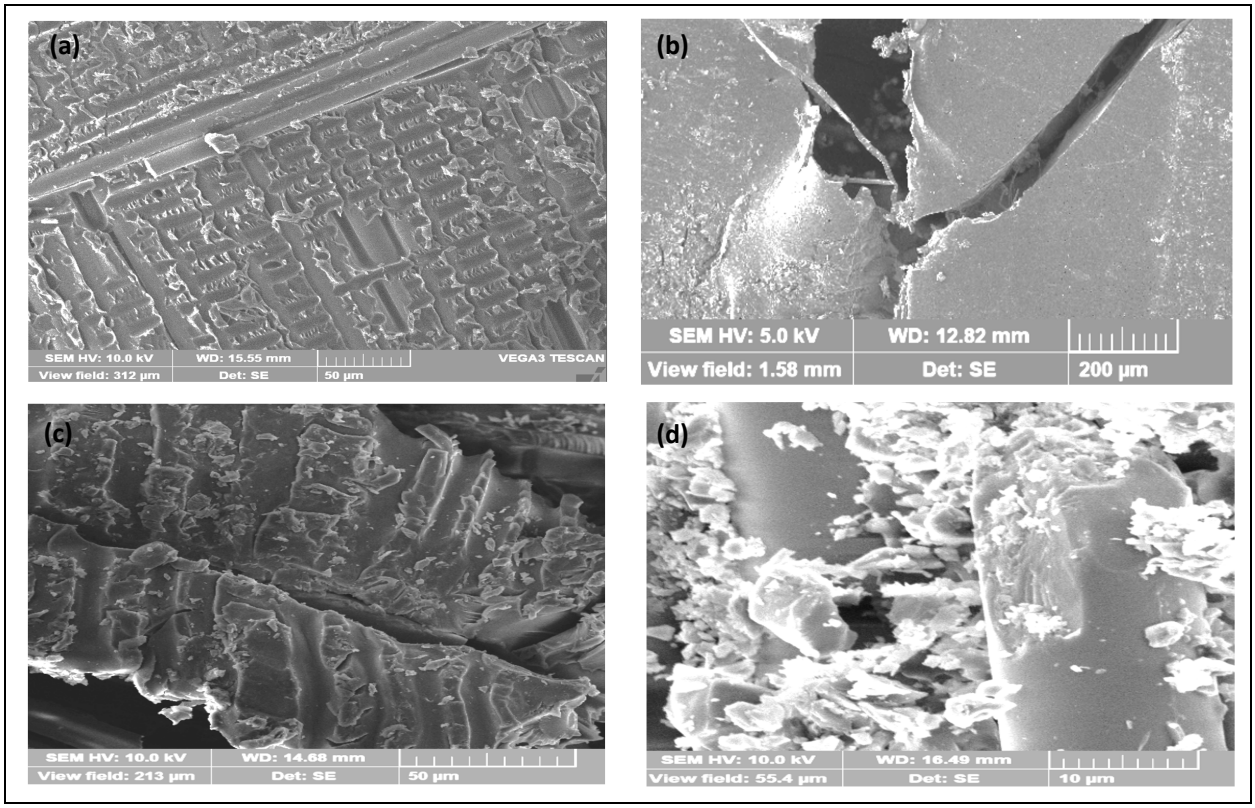


Fig. 4: SEM images of (a) 3%, (b) 6% and (c) 9% (d) 12% of *Camellia sinensis* dust filler incorporated *Vachellia nilotica*/epoxy based composite

The sample with 6% of *Camellia sinensis* dust filler made from *Camellia sinensis* is shown in Fig. 4 (b). As far as the filler distribution is concerned, there is no discernible agglomeration. Their even distribution promotes the effective transfer of stress between the fillers and matrix, improving the mechanical characteristics. Fig. 4 (c) shows the image of the composite fractured surface with 9 v/v% *Camellia sinensis*. This sample also showed a uniform distribution of the filler materials in the matrix. Fillers play a major role in enabling crack-bridging mechanisms, which prevent rupture and are essential for increasing the composite's strength. Fig. 4 (d) shows the formation of filler agglomeration and insufficient bonding between the filler and the matrix when the filler loading surpasses a threshold. Agglomeration causes voids and weak places to occur, as evidenced by the observed loss in the composites tensile, flexural, and impact properties.

3.3. Dynamic Mechanical Analysis

It was observed from the Fig. 5 that the natural filler has a substantial effect on the storage modulus (E'), which measures elastic responsiveness and stiffness. The addition of fillers increases the storage modulus of the composite in both its rubbery and glassy states. Crosslinks prevent intra-chain motion, which increases the stiffness of composite materials. The fillers improve the epoxy matrix's strength at temperatures below T_g, which raises the storage modulus. At temperatures above the T_g, the composite material exhibits a higher storage modulus than the epoxy matrix alone. This suggests that the fillers continue to contribute strength even after the matrix has become flexible.

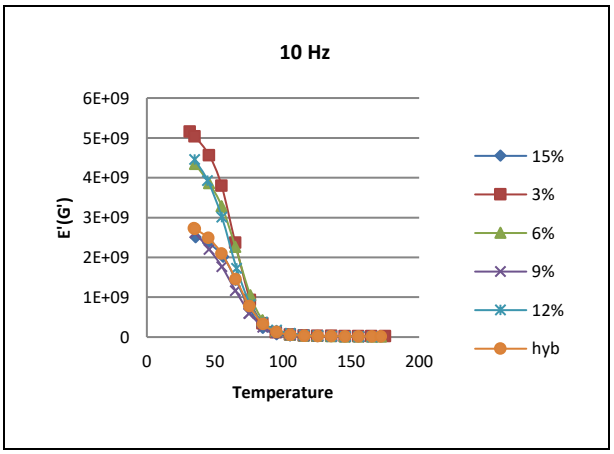


Fig. 5: Storage modulus of *Vachellia nilotica* - epoxy reinforced with *Camellia sinensis* dust filler at 10 Hz frequency

The relationship between tan δ and filler content at a frequency of 10 Hz is given in Fig. 6. The tan δ is the ratio of the storage modulus (E') to the loss modulus (E"). The equilibrium between the heat released by internal molecular friction and the energy retained is depicted in this diagram. The T_g, which indicates the point at which molecular motion becomes more noticeable and the material changes from a glassy to a rubbery state, corresponds to the tan δ peak in composites. The tan δ peak height decreases as filler addition increases to 12 vol.%. The reduction in peak height suggests that the filler particles improve the interface contact between the epoxy matrix and the filler. The composite material exhibits a lower tan δ peak, indicating higher load-bearing efficiency and reduced molecular mobility, whereas the pure epoxy matrix has a higher tan δ peak due to enhanced chain mobility and energy dissipation.

When the filler amount surpasses 12 vol.%, tan δ raised due to agglomeration and microvoid development. Such defects can improve molecular mobility and energy dissipation by preventing filler stress from getting transferred to the matrix. This behavior highlights the importance of adequate filler loading in preserving the mechanical properties of the composite. Incorporation of *Camellia sinensis* dust and *Vachellia nilotica* fillers into the epoxy matrix enhances energy dissipation (by regulating tan δ values) and stiffness, leading to a higher storage modulus. High-performance composites must balance stiffness and damping by ensuring that the fillers are distributed properly and the fibers and the matrix interact to yield these advantages.

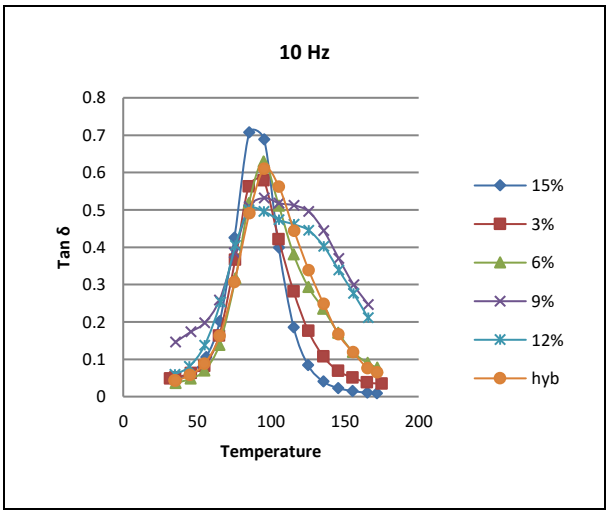


Fig. 6: Effect of Tan δ at 10 Hz frequency on *Camellia sinensis* dust filler reinforced epoxy and *Vachellia nilotica* (VN) composite

3.4. Thermogravimetric Analysis

The TGA results of the *Vachellia nilotica* and *Camellia sinensis* incorporated epoxy is presented in Table 1. According to final, middle and initial degradation temperatures and the residual mass percentages, the addition of filler components increases thermal stability of the composites. The initial degradation temperature of composite materials increases with the addition of *Camellia sinensis* dust

filler, rising from 300 °C for the pure epoxy matrix to 304 °C for composite mixes, including 15% filler.

Table 1. Thermogravimetric analysis (TGA) values

Composition	Degradation Temperature (°C)			Residue (%)
	Stage 1	Stage 2	Stage 3	
Hybrid Epoxy	602	412	300	0.4
3% *Camellia sinensis* dust filler + Hybrid EPOXY	604	413	301	0.6
6% *Camellia sinensis* dust filler + Hybrid EPOXY	606	417	301	0.9
9% *Camellia sinensis* dust filler + Hybrid EPOXY	610	421	302	1.4
12% *Camellia sinensis* dust filler + Hybrid EPOXY	670	432	303	1.6
15% *Camellia sinensis* dust filler + Hybrid EPOXY	677	441	304	1.5

This slight increase illustrates how fillers prolong mechanical failure and enhance the matrix's thermal resilience. The composite's maximum degradation temperature rises with filler concentration, suggesting that fillers enhance thermal stability. The neat epoxy resin had a temperature of 412 °C, while the composite, including 15% filler, had the highest middle decomposition temperature at 441 °C. Fillers function as heat barriers to stop structural degradation. The final decomposition temperature yields comparable results; at 677 °C, the composite with 15% filler exhibited the highest thermal stability. The fillers' ability to create a durable char layer during pyrolysis, which shields the underlying material from heat degradation, is mainly responsible for the improvement. After complete degradation, the residue produced rises according to the filler concentration. The residual in a composite with 12% filler is 1.6%, whereas in a pure epoxy matrix, it is 0.4%. Notably, high residual percentages suggest that composite materials best suited for high-temperature applications are those that resist complete disintegration while maintaining their bulk.

3.5. Dielectric Strength

The experimental results on dielectric strength are shown in Table 2. It can be observed that, at 18.30 kV/mm, the composite with the highest dielectric strength had 3% *Camellia sinensis* dust filler. The Maxwell–Wagner–Sillars approach increases the dielectric strength and induces the polarization effect by immobilizing charge carriers at the filler-matrix interfaces. Above a filler concentration of 3%, the dielectric strength decreases noticeably. At 8.26 kV/mm, the composite with 15% filler has the lowest dielectric strength. The tendency of filler particles to agglomerate at higher concentrations, leading to the creation of microvoids and matrix defects, is the reason for the performance decline. These kinds of defects cause a localized build-up of charge, which raises the risk of dielectric breakdown and lowers performance.

The material can withstand high electric fields without degradation at filler concentrations between 0% and 3% due to the phenomena of charge trapping at the filler-matrix interfaces. Increasing filler content induces mechanical defects, including microcracks and clusters, as well as internal stresses. When an electric field is applied, these defects become regions of decreased dielectric strength. Ultimately, the incorporation of *Camellia sinensis* dust filler improves the epoxy matrix's dielectric strength at lower concentrations. On the other hand, a high filler content causes agglomeration and defects, which lowers the composite's overall dielectric performance. Maintaining an adequate filler loading improves the structural integrity and dielectric characteristics, as indicated by the Maxwell–Wagner–Sillars effect and dielectric strength trends associated with various filler concentrations.

Table 2. Dielectric strength of *Camellia sinensis* filler incorporated *Vachellia nilotica* /epoxy matrix composite

Sample ID	Sea Shells (Vol.%)	Biofiller (Vol.%)	Epoxy Resin (Vol.%)	Dielectric Strength (kV/mm)
1	0	12	88	16.54
2	3	12	85	18.30
3	6	12	82	15.62
4	9	12	79	11.40
5	12	12	76	10.26
6	15	12	73	8.26

5. CONCLUSION

Tensile strength tests show that maximum tensile strength was obtained when the filler loading reached 9 vol.%. However, increasing filler percentages reduce affinity and mechanical robustness, lowering performance. A strong filler-resin interface binding is necessary to maximize energy absorption and impact strength. The SEM images showed uniform filler distribution at 9 vol.%. Dynamic mechanical testing showed that filler in composite materials increases stiffness, energy storage, storage modulus, and glass transition temperature in both glassy and rubbery phases. The damping factor shows that energy dissipation is more at 9 vol.% filler. However, increasing filler concentration reduces matrix-filler interaction and energy dissipation. Thermogravimetric measurements show that the composites with fillers have higher breakdown temperatures, thermal stability, and residual percentages than pure epoxy. Dielectric strength is optimized at low *Camellia sinensis* dust filler concentrations due to well-dispersed particles that promote interfacial polarization. However, more significant filler concentrations might

cause defects, agglomeration, and micro gaps, reducing dielectric performance. *Camellia sinensis* dust filler and *Vachellia nilotica* increase the mechanical, thermal, and dielectric properties of epoxy composites up to a certain filler concentration. When the composite concentration exceeds this level, agglomeration and poor bonding reduce its performance.

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CONFLICTS OF INTEREST

The authors declare that there is no conflict of interest.

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