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Influence of Al_2O_3 - nano filler on the *Vachellia Nilotica* blended hybrid epoxy composites: a comprehensive analysis of mechanical, viscoelastic, and dielectric behavior

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This work presents the influence of Alumina (Al_2O_3) Nano powder on the mechanical, dynamic, and dielectric properties of the hybrid epoxy resin composite material containing *Vachellia nilotica* (VN). The hand lay up method was used to make the composite specimens at different volume fractions of the alumina nano particle filler (3, 6, 9, 12, and 15 v/v%). The *Vachellia nilotica* content was kept at 12 v/v% in all the samples remaining being epoxy resin. The experimental results indicate that the composite with 9 v/v% Al_2O_3 showed better mechanical and dielectric properties when compared to other volume percentages. The epoxy composite with the highest glass transition temperature and storage modulus was also the one containing 9 v/v% nano filler. Furthermore, the dielectric test demonstrated that the addition of the nano filler strengthened the dielectric strength of the composite. The structural morphology of the composite's tensile fracture was investigated using scanning electron microscopy (SEM) to investigate the interaction between the epoxy matrix and the fillers in the hybrid composites.

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1. Introduction

In the last few decades composite materials gained research interest due to necessity in domestic and commercial applications. This new field of study has garnered a lot of interest since it has the potential and provides opportunity to manufacture composite material tailor made for specific applications. The addition of alumina nano fillers to epoxy composites has been the subject of extensive investigation across numerous fields, each concentrating on a different aspect of material development. Several researchers have conducted empirical investigations on the mechanical properties [1-4], thermal properties [5–6], and electrical properties [7–10], and they have reported experimental studies supporting the advantageous application of nano filler integration in epoxy composite materials.

The heat resistance and elastic modulus of the Al_2O_3/epoxy nano-composite shown notable enhancements as reported by Li and Hao [11], particularly the glass transition temperature had been raised by 13.5°C. Mulenga [12] carried out experimental investigation of effect of nano particulate filler (fly ash) on the tensile, flexural and impact properties of a hybrid bio epoxy composite material consisting of sisal fiber-reinforcement. The properties were found to be improved by a percentage of 6.3%, 28%, and 68% respectively due to the inclusion of nano filler. This improvement was ascribed to the mechanical advantages of the fly ash nano-fillers and sisal fibre reinforcing, suggesting the eco-friendly composite's potential for a range of technical uses. According to Wang et al. [13], adding a ternary combination of alumina particles to epoxy composites with varying diameter ratios increased heat conductivity. Achieving a better thermal conductivity of 1.109

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W/m/K, which is a 23%–32% improvement over composites with a single particle size filler, has been demonstrated to require optimization of segregated alumina networks.

According to Wang et al. [14], a sandwiched alumina-epoxy composite with 3 wt% nano-Al_2O_3 added shown a 6.3% increase in breakdown strength over clean epoxy. According to this study, composite materials benefit from the use of alumina nano fillers to enhance their breakdown strength and other characteristics. According to Bommegowda et al. [15], adding alumina nano filler to epoxy composites can have a substantial impact on different mechanical parameters. The study discovered that when 5 wt% alumina was introduced, the composites with the highest loss modulus (2100 MPa) had greater material damping properties. Furthermore, it was proposed that the addition of alumina fillers raised the composites' glass transition temperature, potentially increasing their thermal stability.

The inclusion of TiO_2 nanoparticles in the polymer matrix and its effect on the mechanical properties was reported by Al-Rawi [16] and Kanthavel [17]. Both the researchers reported a significant improvement in the properties due to the inclusion of the nano particulate filler. The study demonstrated that adding nano-filler to the composites increased their natural frequency and damping qualities, highlighting the significance of filler dispersion and interface features in improving composite performance. Othman et al. [18] produced hybrid micro and nano silica filler incorporated epoxy composites, a notable improvement was reported in the mechanical properties. In particular, when both fillers were supplied at a 1:1 weight percentage, Young's modulus increased to 5.39 GPa at 25 wt% loading. Song et al. [19] produced a micro/nano hybrid filler to examine the fracture, tensile and flexural properties of boron carbide-epoxy composites. It was made up of radially oriented BC nanowires as the shell and BC microplatelets as the core, both of which were coated with polyaniline (PANI). The addition of this hybrid filler at a modest concentration of 1 wt% significantly enhanced the composite material's toughness, fracture strain, and elastic modulus. The work demonstrated the application of multiscale reinforcement design to produce durable, lightweight, and robust materials with improved mechanical performance and load-carrying efficiency [20].

A detailed analysis of the impacts of fillers on characteristics including glass transition temperature, damping factor, loss modulus, and storage modulus was done in order to throw light on the complimentary effects of fillers and fabric combinations. The study revealed percentage increases in both storage modulus and loss modulus; the GC2 composite had a 49% increase in storage modulus and the GK2 composite a 38% increase in loss modulus when compared to its counterparts without filler. Hu et al.'s study [21] on epoxy adhesives modified by inorganic fillers found that an impact strength of 28.43 KJ/m^2 was obtained at 60% micro alumina concentration, which had the best toughening effect. This outcome highlights the significance of an alumina nanofiller in raising the impact strength of hybrid epoxy composite materials, which raises the materials' mechanical attributes. The study by Balakrishnan [22] demonstrated the effect of nano particulate filler loading on the mechanical properties of epoxy composites. It highlighted that a 2.5 wt% filler content enhanced tensile strength, flexural strength, and hardness while a 10 wt% filler content boosts compressive strength and impact strength. Nimbagal, V. et al. [23] investigated the mechanical and fracture properties of carbon nanofiber/short carbon fibre epoxy composites. It was reported that the hybrid nano composites' tensile strength was 17.18–25.72% greater than the CNF-reinforced nano composites'. While the impact strength had significantly improved by 39.87–97.05%, the flexural strength demonstrated a notable improvement of 39.24–44.07%. These improvements in mechanical characteristics showed that CNFs and SCFs worked together in the epoxy matrix. Interestingly, the hybrid nanocomposites' fracture toughness had also increased. Compared to the neat epoxy-PLA composite, there was a 37.93–38.77% improvement in fracture toughness. According to the literature, limited studies had been reported on the influence of nanofiller on the hybrid composite materials. This work involves the fabrication of novel hybrid composites incorporated with nano particulate filler (Alumina) and an experimental investigation of mechanical, thermal, and dielectric characteristics.

2. Materials and methods

2.1. Materials

Epoxy resin, commercially known as Araldite LY 556 and hardener (HY951) were used as base matrix material for this experimental study. The materials were purchased from Sakthi Fibres, Chennai, India, the Epoxy and hardener were mixed in a 10:1 ratio. Alumina nanoparticles which have an average size of 50 nm, was procured from Sakthi Fibres, Chennai, India. These nanoparticles' remarkable mechanical qualities and thermal stability make them perfect for strengthening the epoxy matrix. Powdered *Vachellia nilotica* (Babul) with a particle size of around 100 µm was obtained from Vimal Enterprises in Kanchipuram, India, for use as a natural filler. Combining *Vachellia nilotica* with alumina nanoparticles may enhance the mechanical properties of the composite because it is a readily available and biodegradable material.

2.2. Testing methods

The produced composite samples were evaluated for their dielectric, mechanical, and dynamic mechanical properties using standard ASTM tests. Mechanical characteristics including modulus and tensile strength were assessed using ASTM D638 on an Instron 5567 universal testing machine running at a crosshead speed of 2 mm/min. Flexural characteristics were evaluated using an Instron machine that was set up with a three-point bending fixture and ran at 10mm/min in compliance with ASTM D790. Using ASTM D256 the Charpy impact tester (Ceast Resil Impactor, Italy) was utilized to assess impact strength. Five samples are tested, and the average is considered for analysis.

Viscoelastic properties such as storage modulus (E') and loss modulus (E") were measured per ASTM E1640 on a (Model: SEIKODMAI-DMSC 6100) in tensile mode at 1 Hz frequency and with a steady heating rate of 5°C/min. The damping factor, Tan δ, was calculated by dividing the storage modulus by the loss modulus. The maximum voltage at which a material disintegrates in known as dielectric strength. In this work CEAST 6135 a breakdown voltage analyzer, with a maximum operating voltage of 60 kV, is used to determine the breakdown voltage of epoxy resin with *Vachellia nilotica* gum composites in compliance with ASTM D 149 requirements. A round specimen of 80 mm diameter and 3 mm thickness was used in the testing device in which the voltage was raised at 100V per min. The dielectric strength was calculated as the mean of the five examined identical specimens.

3. Results and discussion

3.1. Tensile strength

Figures 1 through 3 illustrate the effect of nano alumina on the tensile, flexural, and impact strength. of hybrid matrix epoxy composites. Plot of tensile strength as shown in fig 1 indicates a minor improvement with nano filler volumes up to 9%; however, as filler content rises above 9%, tensile strength starts to decline, as seen in Figure 1. Furthermore, the plot demonstrates that the tensile modulus falls after decreasing linearly up to 9 v/v% nano filler. When more natural filler powder and nano filler were combined with resin, poor bonding and a decrease in mechanical strength were observed. Additionally, it has been noted that the dispersion of the natural filler powder, which is made up of broken and crushed particles, activates the strengthening mechanisms of the epoxy matrix. The advantages of natural filler materials are rupture arresting and crack stapling, which can boost the strength of hybrid composites.

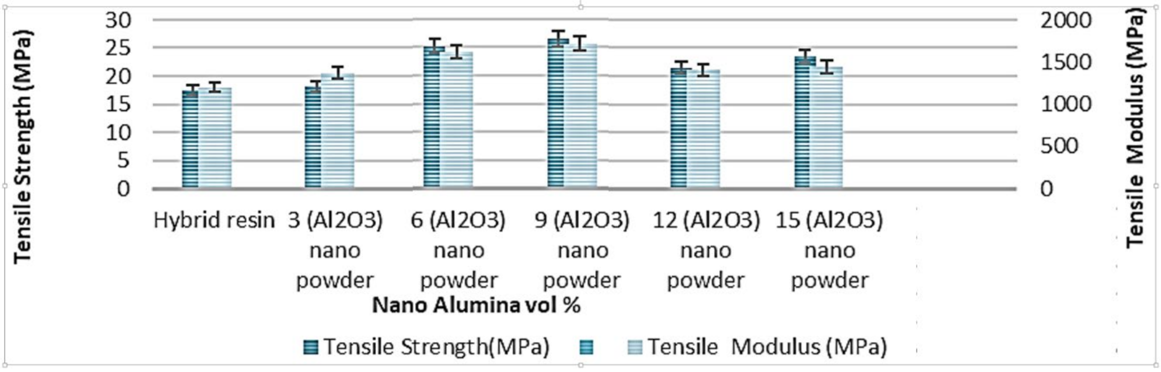


Fig. 1. Tensile strength of nano Alumina - Vachellia nilotica /epoxy matrix composite.

As seen in Figure 2, the addition of nano filler and reinforcement made of *Vachellia nilotica* has changed the composite's flexural properties. Flexural strength and modulus showed a similar trend as that of tensile properties. Flexural strength increases at 9 v/v% of the composite material; however, flexural strength decreases at filler powder volumes above 9%. It is evident from a comparison of the composite's flexural and tensile strengths that the fibre alignment in the outer layer surface is what gives the composite its higher flexural strength.

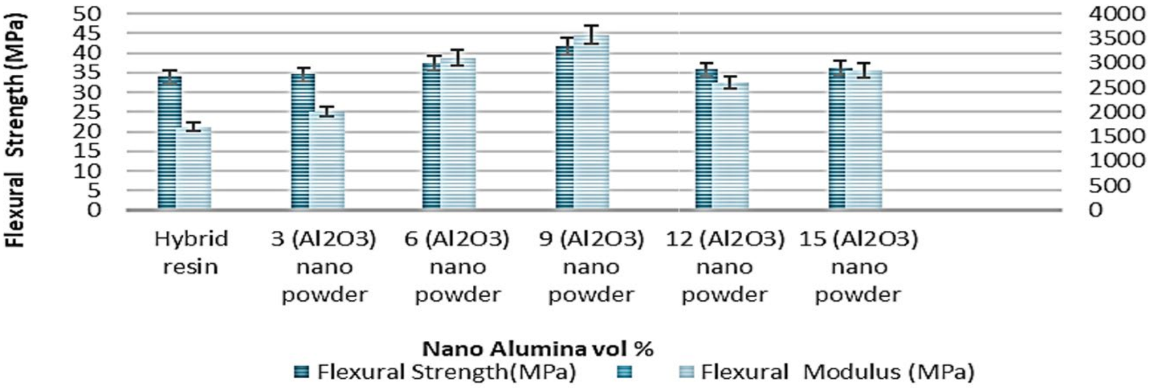


Fig. 2. Flexural strength of Vachellia nilotica /epoxy matrix composite.

Nanoparticles have an impact on the hybrid matrix epoxy composite's impact strength, as Figure 3 illustrates. The composite material's ability to absorb energy explains why it has a higher impact strength. Epoxy resin could act as a fracture cover by combining with nano filler, developing into a more intricate pattern and absorbing more energy.

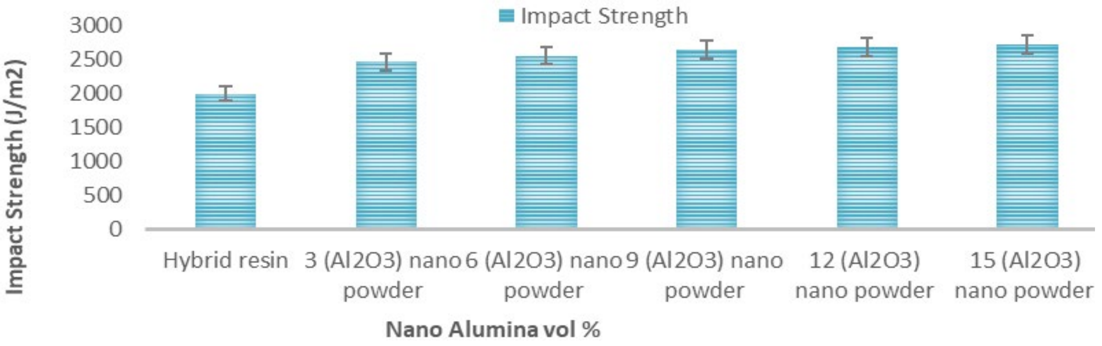
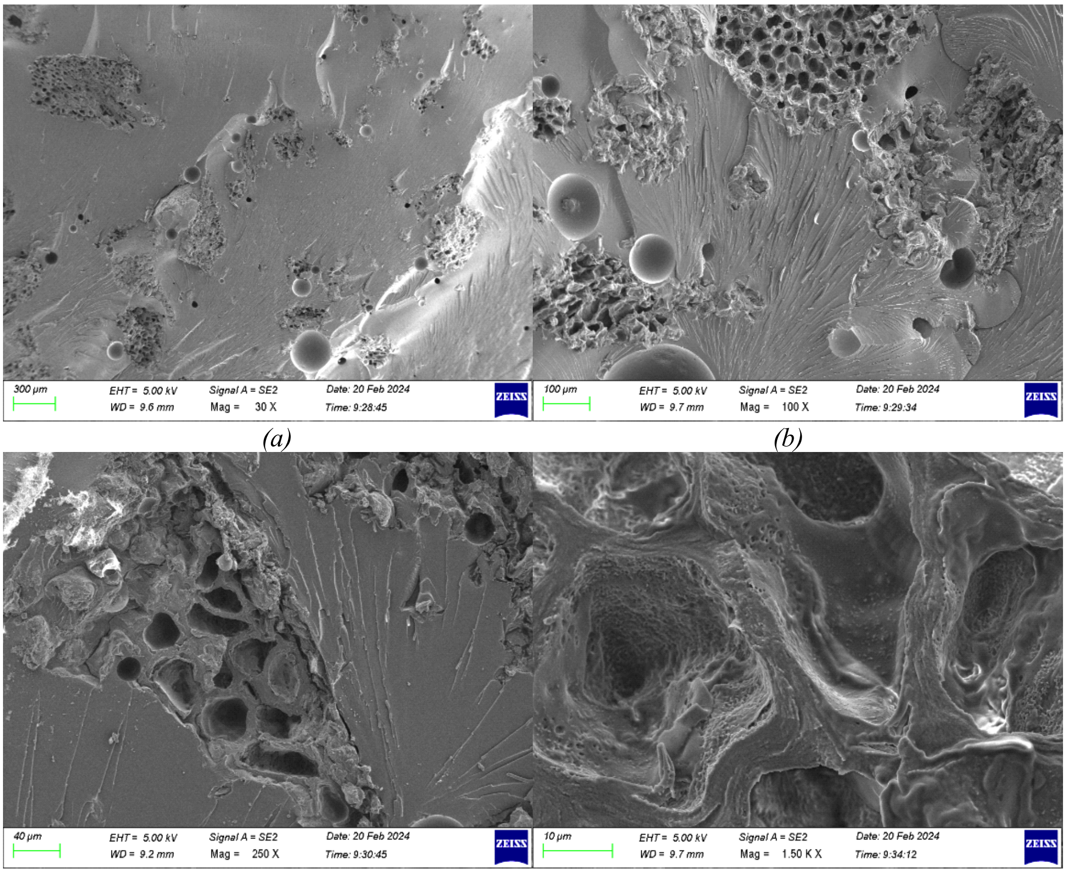


Fig. 3. Impact strength of Vachellia nilotica (VN)/epoxy matrix composite.

3.2. SEM analysis

The matrix-filler interaction and surface adhesion between the constituent materials of the tensile fractured specimen were examined using SEM. The micrographs resulting from the variation using nano filler *Vachellia nilotica* filler/epoxy composite are shown in Fig. 4(a)–(d). As seen in Figure 4(a), there is very little Al_2O_3 and no agglomeration from the epoxy/natural filler composite. The uniform dispersion of the nano filler at 9 v/v% and the natural filler with polymer are responsible for the equal distribution of the imposed load on the composite, as demonstrated by the SEM micrograph in Figures 4b and 4c. The filler agglomeration and coalescence between the filler and matrix in the composite with 12 vol% filler material demonstrated poor bonding when compared to other composites of different compositions. The composite in figure 4(d) made this very evident.



(a) *(b)*

(c) *(d)*

Fig. 4. SEM images of (a)3%, (b)6% and (c)9% (d) 12% of Al_2O_3 nano filler incorporated Vachellia nilotica (VN)/epoxy based composite.

3.3. Visco - elastic analysis

3.3.1. Storage modulus (E')

The viscoelastic property analysis in as shown in figure 5 illustrates the effect of temperature and filler loading on the storage modulus at a frequency of 1 Hz. The results suggest that the nano filler increases the composite material's ability to absorb the energy under cyclic load. As Figure 5 shows, the basic epoxy resin has a lower storage modulus and is hence stiffer. The higher glass transition temperature (Tg) of 100 °C of composite materials is attributed to the free molecular mobility of the epoxy resin polymer chain. Studies have demonstrated that the addition of filler to the polymer matrix increases the stiffness of the composite material. Both the solidus and liquidus zones exhibit the effect of filler material as the storage modulus increases. Studies show that adding a nano filler to the epoxy matrix improves the stiffness of the brittle and ductile portions of hybrid composite materials.

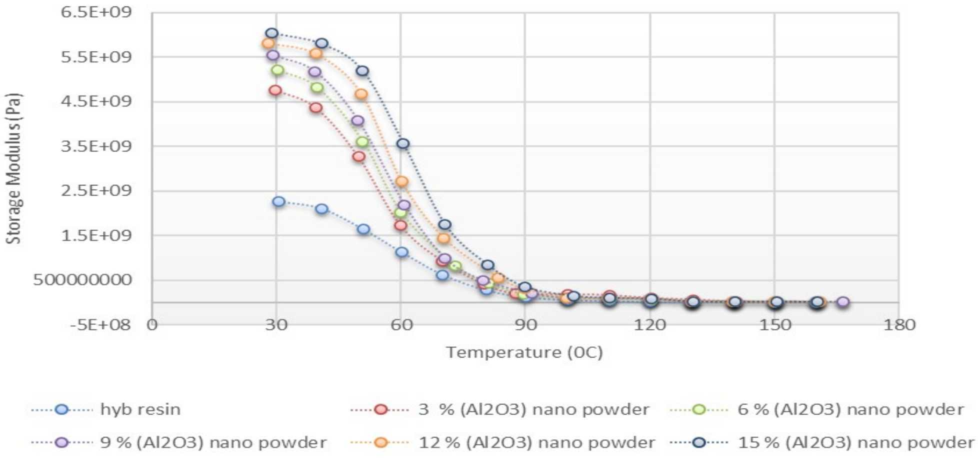


Fig. 5. Storage modulus of epoxy and Vachellia nilotica at 1 Hz frequency

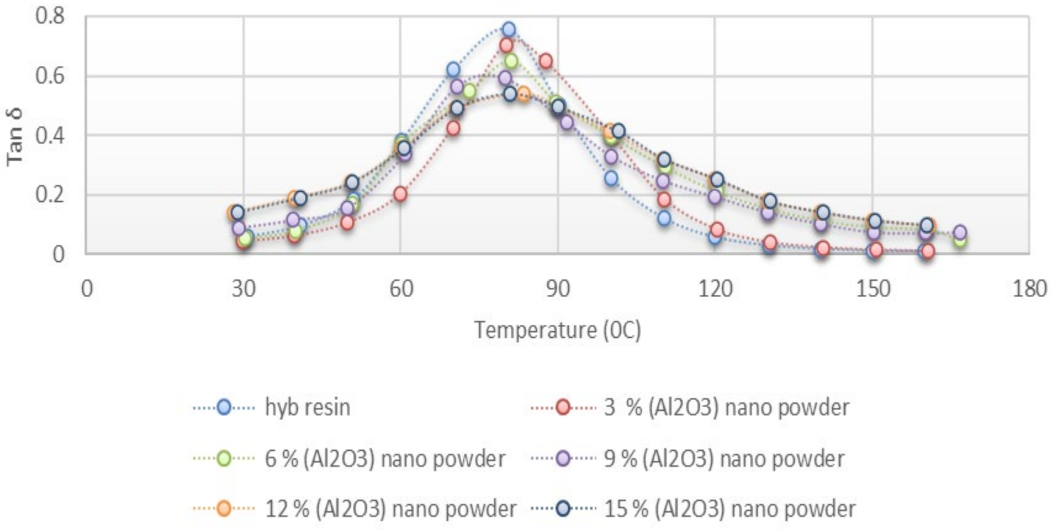


Fig.6. Effect of Tan δ at 1 Hz frequency on epoxy and Vachellia nilotica (VN) composite.

3.3.2. Damping factor

Damping factor was calculated as the material's ratio of storage modulus to loss modulus, which is an indication of degree of molecular mobility in the polymer chain as well as the energy dissipation ability of composite material under cyclic load.

The damping factor of the Al_2O_3 nano filler combined with the epoxy filler and the composite material made of *Vachellia nilotica* (VN) is displayed in Figure 6. It was discovered that the matrix-filler interaction increases as the volume fraction of filler loading decreases. The enhanced contact causes the composite material to dissipate more energy. Conversely, energy dissipation diminishes when filler content increases (over 12% volume percent), as Figure 6 illustrates. Lower peak height indicates greater interfacial bonding in between the hybrid epoxy resin matrix and nano filler.

3.4. Thermogravimetric analysis

Table 1 shows the thermogravimetric analysis (TGA) curves and the basic epoxy resin used in the composites. The hybrid epoxy with nano filler shows better thermal stability than a hybrid epoxy matrix. The results show that the addition of nano filler improves the thermal stability of the hybrid epoxy matrix.

Table 1. Thermogravimetric Analysis (TGA) values.

Composition	**Degradation temperature (°C)**			**Residue (%)**
	FDT	**MDT**	**IDT**	
Epoxy + 12 Bio filler	402	312	200	0.2
3% Al_2O_3 + Hybrid EPOXY	404	313	201	0.4
6% Al_2O_3 + Hybrid EPOXY	406	317	201	0.8
9% Al_2O_3 + Hybrid EPOXY	410	321	202	1.1
12% Al_2O_3 + Hybrid EPOXY	470	332	203	1.2
15% Al_2O_3 + Hybrid EPOXY	477	341	204	1.5

The filler components in the composite account for the data's observed improvement in TGA levels. The addition of nano filler enhances the epoxy matrix's resistance to heat. Table 1 displays the natural state thermal stability of the epoxy matrix at 200°C. Each specimen shows that residues begin to form and all combustible elements are eliminated completely within the temperature range of (402-477) °C. The *Vachellia nilotica* incorporated hybrid epoxy composite was stable up to 12% VN, leaving 1.2% of residual nano filler it swiftly degrades below this temperature, leaving 0.3% of residue. This shows that the experimental thermal stability of alumina nano filler incorporated *Vachellia nilotica* epoxy composite is greater than neat epoxy resin as found in the literature.

3.5. Dielectric strength

Nano filler is attributed in improving the dielectric strength of the *Vachellia nilotica* incorporated epoxy matrix. Table 2 displays the dielectric breakdown voltage values of epoxy and nano filler loading on the epoxy composites. The greatest dielectric strength of the epoxy/*Vachellia nilotica* resin sample was 15.30 KV/mm. The hybrid composite containing 3 vol.% nano alumina particle natural filler composite has the best dielectric strength among the others. Nanoparticles are evenly distributed throughout the epoxy matrix at low alumina concentrations (0% and 3%), which enhances interfacial polarisation. The Maxwell-Wagner-Sillars (MWS) effect supports the increase in dielectric strength induced by effective charge trapping at interfaces as a result of this polarisation. Above 3%, there is a tendency for particles to coalesce into larger groups. This agglomeration produces microvoids and flaws, which lead to a localised buildup of charge and a loss in dielectric strength. By creating internal stress, a higher filler content erodes the composite's mechanical strength. Clusters and microcracks occur with high filler loadings, lowering the dielectric performance. Mechanical defects may serve as triggers for electrical failure in the presence of powerful electric fields.

Table 2. Dielectric strength of Alumina nano filler incorporated Vachellia nilotica /epoxy matrix composite.

Sample ID	**Volume Fraction of Alumina (%)**	**Volume Fraction of Bio Filler (%)**	**Volume Fraction of Epoxy Resin (%)**	**Dielectric Strength (kV/mm)**
1	0	12	88	13.54
2	3	12	85	15.30
3	6	12	82	12.62
4	9	12	79	10.40
5	12	12	76	9.26
6	15	12	73	7.26

Conclusions

This work examined the mechanical, dielectric, and thermal properties of hybrid matrix epoxy composites enhanced by *Vachellia nilotica* gum and Al_2O_3 nano filler varying in volume percentage. Tensile strength measurements initially indicate an increase in filler up to 9 vol%; however, at higher filler percentages, there is a drop due to insufficient bonding and mechanical strength loss. Strong interfacial adhesion between the resin and filler boosts impact strength and improves the material's energy absorption capacity. At 9 vol%, the SEM analysis showed uniform filler dispersion. According to dynamic mechanical studies, the addition of filler improves the stiffness and energy storage in both the glassy and rubbery stages of composite materials, hence increasing their storage modulus and glass transition temperature (Tg).

According to the damping factor study, energy dissipation reaches its peak at 9 vol% filler, and as filler concentrations rise, less matrix-filler interaction causes dissipation to decrease. A thermogravimetric investigation shows that, in comparison to pure epoxy, composites including fillers show greater thermal stability, as well as superior degradation temperatures and residue percentages. At low alumina concentrations (3 vol%, to be exact), dielectric strength is maximised due to well-dispersed nanoparticles that improve interfacial polarisation. Higher filler concentrations, however, result in flaws, agglomeration, and microvoids that lower dielectric performance. Overall, adding nano fillers and VN to the epoxy matrix enhances the composite's mechanical, thermal, and dielectric properties up to an optimal filler concentration; after that, agglomeration and poor bonding result in a decrease in performance.

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