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SYNERGISTIC EFFECTS OF HYBRID MONTMORILLONITE AND GRAPHENE NANOPLATELETS ON THE MECHANICAL, THERMAL, AND STRUCTURAL PROPERTIES OF EPOXIDIZED PALM OIL/EPOXY COMPOSITES

Vegetable oils (VOs) are increasingly recognized as promising renewable alternatives to traditional epoxy resins. However, their relatively modest mechanical and thermal properties often hinder broader application. To overcome these limitations, this study focused on enhancing the performance of epoxy/epoxidized palm oil (EEPO) through a synergistic reinforcement strategy involving hybrid nanofillers. The hybrid filler systems were developed by combining a fixed amount of graphene nanoplatelets (GNP) at 0.5 phr with varying loadings of montmorillonite (MMT) at 1.0, 1.5, and 2.0 phr. The hybrid fillers were incorporated into the developed resin system to evaluate their synergistic effects on the overall performance of the composite. Comprehensive characterization, including X-ray diffraction (XRD), scanning electron microscopy (SEM), flexural testing, and thermal analyses such as differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA), was conducted to examine the structural, mechanical, and thermal behavior of the resulting composites. The hybrid composite containing EEPO/GNP-MMT (0.5:1.0) achieved the highest performance, with a 37.7% increase in flexural strength (86.31 MPa), a 9.5% increase in flexural modulus (2.87 GPa), and a 20.6% increase in glass transition temperature (67.79°C). TGA results revealed improved thermal stability due to the enhanced barrier effects and the filler-matrix interfacial bonding. These results highlight the synergistic role of hybrid MMT and GNP fillers in producing sustainable, high-performance epoxy composites for structural applications, protective coatings, and bio-based panels.

Keywords: Epoxy-epoxidized palm oil blend; graphene nanoplatelets (GNPs); montmorillonite (MMT); mechanical and thermal properties; hybrid nanofillers

1. Introduction

Research is currently focused on the development of bio-based thermosets as alternatives to petroleum resins such as epoxy. Vegetable oils (VOs) have emerged as attractive candidates because of their low cost, availability, and versatility [1]. The triglycerides in vegetable oils can be modified to produce resins with epoxy functionality. Malaysia, as the second-largest producer of palm oil after Indonesia, processed 19.1 million tonnes of crude palm oil in 2020, highlighting the significant potential of palm oil as a bio-source [2]. However, VOs have chemical structural constraints that hinder the production of high-quality resins comparable to petroleum resins. Therefore, blending vegetable oils with petroleum-based epoxies is a common approach to achieve synergistic properties. The incorporation of natural fillers such as flax, basalt, and glass, and nanofillers such as carbon nanotubes (CNTs), nanocarbon, montmorillonite (MMT), graphene, and aluminium, have been widely reported to enhance the mechanical and thermal properties of epoxy resins [3,4]. Graphene (a two-dimensional, sp^2-hybridized carbon nanomaterial) is well known for its superior thermal, mechanical, and electrical properties, making it an effective nanofiller in polymer composites [5]. Montmorillonite (MMT), on the other hand, has a layered crystal structure and can be organically modified (e.g., Cloisite 30B) to improve compatibility and dispersion within polymer matrices [6].

The nanofiller content has a significant impact on the reinforcement of the composite. Gholami et al. [7] reported that the addition of 0.3 wt.% multi-walled carbon nanotubes (MWCNTs) and graphene oxide nanoplatelets (GONPs) increased strength, whereas higher loadings (0.5 wt.%) led to aggregation and reduced performance. Singh et al. [8] observed improved tensile and impact strength with silica nanoparticles up to 2 wt.%, followed by a decline at higher concentrations. The combination of two distinct nanofillers can overcome dispersion challenges and balance property trade-offs, resulting in high-performance composites.

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For example, Charitos et al. [9] showed that the combination of graphene oxide (GO) and CNTs improved electrical conductivity, while Daraei and Ghaemi [10] reported an improvement in membrane properties through the use of Cloisite 15A and 30B in polyethersulfone (PES). These findings suggest that the strategic selection of complementary nanofillers can produce synergistic effects, although the extent of such benefits depends on filler morphology, dimensionality, and interaction with the polymer matrix.

While most previous research has focused on combining nanofillers with distinct shapes or dimensionalities, the synergistic potential of nanofillers with comparable morphologies remains less explored. In this context, Mohammed et al. [5] found that the hybridization of montmorillonite (MMT) and graphene nanoplatelets (GNPs) in carbon fibre-reinforced composites (CFRCs) resulted in balanced improvements in both mechanical and thermal performance. This indicates that even nanofillers with similar geometrical characteristics can exhibit cooperative reinforcement effects when properly dispersed. Such findings are particularly relevant to epoxy-epoxidized palm oil (EEPO) systems, which in previous studies exhibited reduced stiffness and lower thermal stability due to the flexible molecular structure of the epoxidized palm oil (EPO) component [11]. Therefore, the present study employs a hybrid nanofiller strategy combining MMT and GNPs to enhance the mechanical and thermal performance of EEPO resins. To date, this approach has not been systematically investigated, and the findings are expected to advance understanding of hybrid nanofiller interactions in EEPO-based composite systems.

2. Experiment

The control sample, consisting of epoxy-epoxidized palm oil (EEPO) resin without nanofillers, was prepared using a conventional direct blending method. A diglycidyl ether of bisphenol A (DGEBA)-based epoxy resin with an equivalent weight of 182 to 192 g/eq was used in combination with the curing agent Jeffamine® 905-3S. Epoxidized palm oil (EPO), containing approximately 1.5 epoxide groups per triglyceride, was incorporated at 10 wt.%. The epoxy-to-EPO ratio was maintained at 90:10, in line with previous studies indicating that higher EPO content can deteriorate mechanical and thermal performance. The epoxy resin was heated to 55°C, after which EPO was gradually added under continuous stirring until a homogeneous mixture was obtained. The blend was then cooled to room temperature before adding the curing agent. The prepared mixture was poured into a closed mould and cured at 110°C for two hours. The cured EEPO composite was cut into suitable specimen dimensions for subsequent testing and analysis.

For the nanocomposite samples, a solvent-assisted dispersion technique was employed. Graphene nanoplatelets (GNPs) and montmorillonite (MMT) were dispersed in acetone and sonicated for six hours to achieve uniform distribution. The resulting nanofiller suspension was introduced into the epoxy resin, and the mixture was heated to 80°C with continuous stirring to completely evaporate the acetone. After acetone removal, the mixture was cooled to approximately 55°C, and EPO was added with thorough stirring to ensure uniform distribution. Once cooled to room temperature, the curing agent was incorporated. The final formulation was poured into an aluminium mould and allowed to cure under ambient conditions. Hybrid nanocomposites containing both GNPs and MMT were fabricated using the same procedure, encompassing nanofiller dispersion, resin blending, and curing. The GNP-to-MMT ratios of all formulations are presented in TABLE 1, while the overall fabrication process of the EEPO/MMT-GNP nanocomposites is illustrated in Fig. 1.

For testing and characterization, X-ray diffraction (XRD) was carried out using a Rigaku SmartLab diffractometer to study

TABLE 1

Formulations of EEPO/GNP-MMT Nanocomposites

Formulation (%)	Epoxy (wt.%)	Epoxidized Palm Oil (wt.%)	Graphene nanoplatelets (GNP) (phr)	Nanoclay MMT (Cloisite 30B) (phr)
EEPO	90	10	—	—
EEPO/GNP-MMT (0.5:0)	90	10	0.5	0
EEPO/GNP-MMT (0.5:1.0)	90	10	0.5	1.0
EEPO/GNP-MMT (0.5:1.5)	90	10	0.5	1.5
EEPO/GNP-MMT (0.5:2.0)	90	10	0.5	2.0

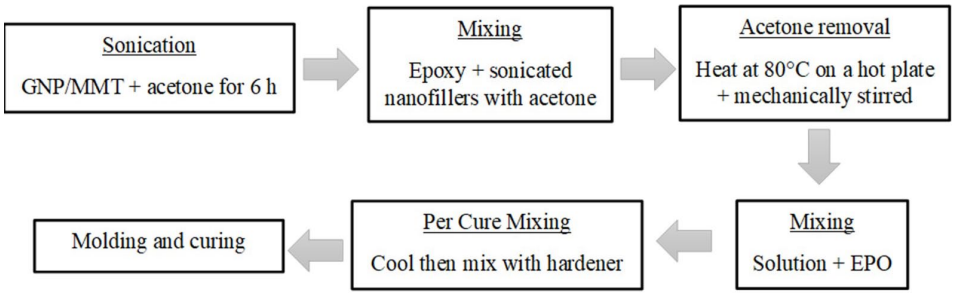

Fig. 1. Fabrication process of the EEPO/GNP-MMT nanocomposites

nanofiller dispersion within a 2θ range of 3°-60°. Mechanical testing was conducted on a Lloyd Universal Testing Machine (Model 4400, Lloyd Instruments, UK) with a maximum load capacity of 10 kN, using the three-point bending method (ASTM D790). Tests were performed at a crosshead speed of 2.8 mm/min with a span length of 50 mm. Specimens of 12.7×1.27×0.5 cm^3 were tested, with seven replicates per formulation, and average flexural strength and modulus were calculated. Thermal properties were evaluated by thermogravimetric analysis (TGA, PerkinElmer TGA 7) on approximately 10 mg samples heated from 30-600°C at 10°C/min under nitrogen. Differential scanning calorimetry (DSC) was conducted under nitrogen (50 mL/min) on 5-12 mg dried samples sealed in aluminium pans. The samples were heated from room temperature to 300°C at 5°C/min under four different heating programs to determine curing kinetics. Cross-sectional morphology was examined using a TESCAN VEGA Compact scanning electron microscope (SEM) after gold sputter-coating, at an accelerating voltage of 20 kV.

3. Results and discussion

3.1. X-Ray Diffraction (XRD) analysis

X-ray diffraction (XRD) analysis was conducted to investigate the structural organization and dispersion behavior of the hybrid fillers within the cured resin matrix. Fig. 2 shows the XRD patterns of graphene nanoplatelets (GNPs), montmorillonite (MMT), and EEPO resin blends with GNP-MMT hybrids at various ratios. The pristine MMT exhibited a sharp basal reflection at 2θ = 4.85°, corresponding to an interlayer spacing of 1.82 nm, which represents the initial gallery distance of the clay layers in the absence of other materials.

After hybridization with GNP and incorporation into the EEPO matrix, the characteristic MMT peaks shifted to 3.81° (2.32 nm), 3.49° (2.53 nm), and 3.89° (2.27 nm) for EEPO/GNP-MMT (0.5:1.0), EEPO/GNP-MMT (0.5:1.5) and EEPO/GNP-MMT (0.5:2.0), respectively. This shift to lower diffraction angles indicates a clear expansion of the MMT interlayer spacing, suggesting the intercalation of polymer chains or other hybrid components into the silicate galleries; the largest expansion is observed for EEPO/GNP-MMT (0.5:1.5).

No distinct GNP diffraction peaks were detected in any of the hybrid composites, indicating that the GNP sheets were fully dispersed within the polymer matrix. The absence of these peaks, combined with the observed d-spacing increase for MMT, strongly suggests that GNPs play a synergistic role in improving MMT dispersion [12]. It is likely that the GNP sheets act as both physical spacers and nucleation sites, enabling polymer penetration between MMT platelets and promoting their separation [12]. The absence of GNP peaks, together with the increased MMT d-spacing, supports the interpretation that GNPs assist MMT delamination, likely by acting as spacers and promoting polymer infiltration between platelets. In particular, the two-dimensional sheet shape and high aspect ratio of GNPs allow these particles to be inserted between the MMT layers, thus preventing the rearrangement of the layers through physical effects and creating sufficient steric hindrance for effective separation. This "spacer effect" phenomenon aids the MMT intercalation process by widening the distance between the galleries, which in turn facilitates the penetration and binding of polymer chains in the interlayer space, resulting in a more uniform dispersion of the filler in the composite matrix. Furthermore, the good interfacial compatibility between MMT and GNPs may result in strong interactions with the polymer matrix through hydrogen bonding and van der Waals forces, thus strengthening the interphase adhesion and promoting more effective clay delamination and dispersion. [13].

During melt blending, collisions between GNP and MMT particles could generate localized shear forces and mechanical stresses, contributing to the further separation of MMT layers. The hybrid morphology resulting from this process, characterized by well-dispersed GNPs and increased MMT interlayer spacing, provides strong evidence of improved filler dispersion. Such a uniformly dispersed filler network, as confirmed by XRD, is known to enhance load-transfer efficiency and restrict polymer chain mobility – mechanisms commonly associated with improved mechanical and thermal performance [14], as discussed in subsequent sections.

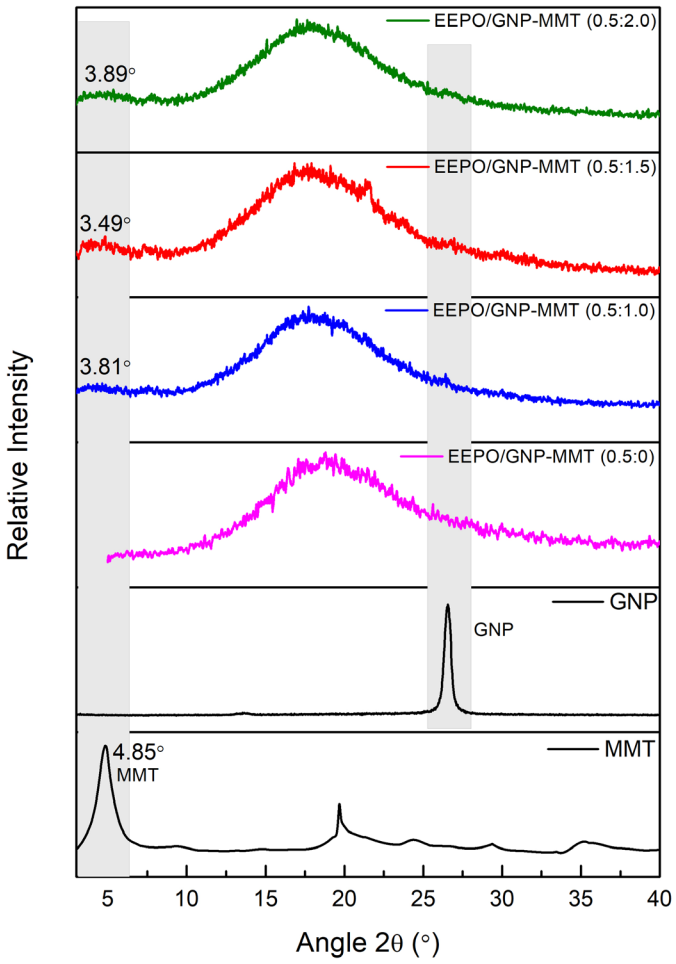

Fig. 2. XRD patterns of EEPO incorporated with GNP:MMT hybrid of different ratios

3.2. Mechanical properties

3.2.1. Flexural properties

Figs. 3 and 4 illustrate the variation in flexural strength and modulus of EEPO composites incorporating a fixed GNP content of 0.5 phr with different MMT loadings (0, 1.0, 1.5, and 2.0 phr). A clear performance trend was observed across the formulations. Compared to the neat resin, the addition of 0.5 phr GNP alone increased the flexural strength from 62.67 MPa to 72.66 MPa, representing an improvement of 15.9%, while the modulus increased slightly from 2.74 GPa to 2.76 GPa. This initial enhancement can be attributed to the high aspect ratio of GNP, which facilitates efficient stress transfer across the polymer matrix and promotes stronger filler–matrix interfacial bonding [15,16]. Further improvement was observed upon the incorporation of 1.0 phr MMT together with 0.5 phr GNP, where the flexural strength reached 72.83 MPa and the modulus increased to 2.78 GPa. This indicates that at low MMT loading, the clay platelets restrict polymer chain mobility while complementing the reinforcing effect of GNP, leading to a moderate increase in stiffness.

The optimum performance was achieved at EEPO/GNP-MMT (0.5:1.5), where the flexural strength peaked at 86.31 MPa and the modulus reached 2.86 GPa. This significant enhancement is attributed to the synergistic interaction between GNP and MMT: GNP facilitates the uniform dispersion of MMT platelets, while MMT provides a rigid framework that effectively restricts polymer chain motion [13,17]. The resulting homogeneous filler distribution maximizes stress-transfer efficiency and improves the overall stiffness of the composite. Similar synergistic effects have also been reported in biopolymer-based hybrid systems such as PLA/GNP–MMT and PVA/GNP–MMT, where the uniform dispersion of GNPs and extensive exfoliation of MMT strengthen the interfacial interactions and enhance the flexural properties through hydrogen bond formation and van der Waals attraction [18,19]. Tavares et al. [18] reported a 22% increase in flexural strength for biopolymer nanocomposites containing GNPs, resulting from more efficient stress transfer and optimal filler dispersion. Meanwhile, Velo et al. [19] also found a comparable increase in bio-resin-based graphene oxide-MMT hybrid systems.

However, in this study, at EEPO/GNP-MMT (0.5:2.0), both flexural strength (62.53 MPa) and modulus (2.75 GPa) decreased markedly. This deterioration is likely due to the aggregation and rearrangement of MMT platelets at higher loadings, which disrupt uniform filler dispersion and weaken polymer-filler interfacial interactions, thereby reducing stress transfer [20]. This trend is consistent with the XRD results, which show the largest interlayer spacing (d-spacing) at EEPO/GNP-MMT (0.5:1.5) – indicating the highest degree of filler separation and better dispersion. At EEPO/GNP-MMT (0.5:2.0), the reduction in d-spacing suggests closer platelet packing, which may hinder effective dispersion within the polymer matrix. These results agree with previous studies on hybrid filler systems, where optimal reinforcement typically occurs at moderate filler loadings before aggregation effects dominate [20].

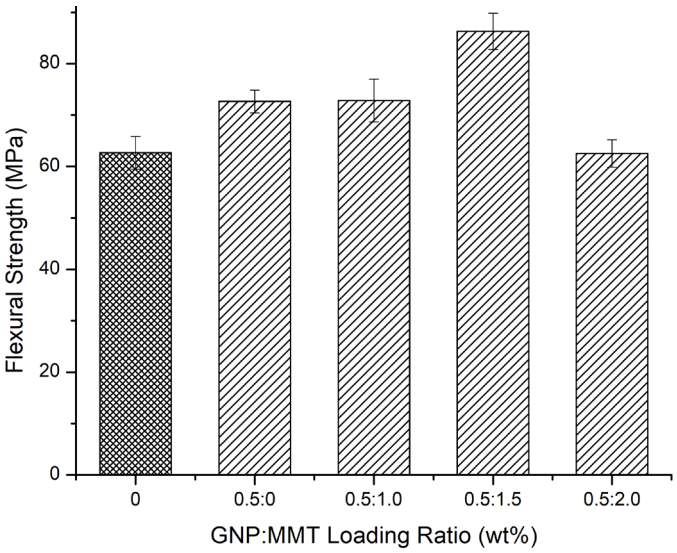

Fig. 3. Flexural strength of EEPO with different ratios of GNP:MMT

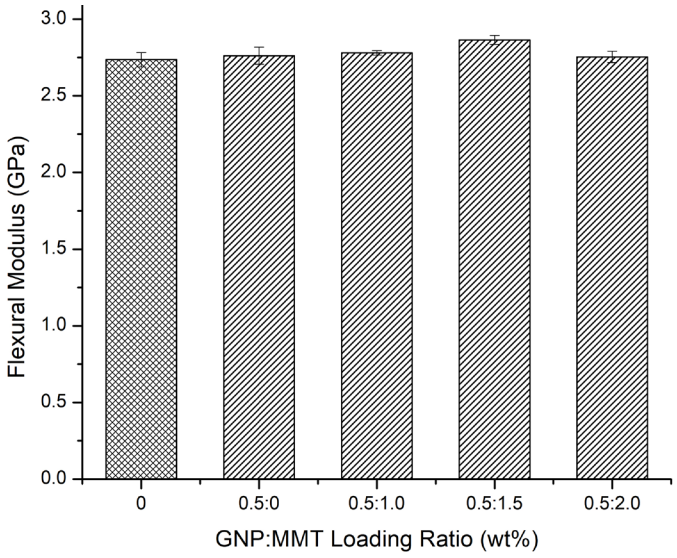

Fig. 4. Flexural modulus of EEPO with different ratios of GNP:MMT

3.3. Thermal properties

3.3.1. Thermal Gravimetric Analysis (TGA)

TGA analysis was performed to evaluate the effect of GNP–MMT hybrid incorporation on the thermal stability and degradation profile of the EEPO resin. Fig. 5 and TABLE 2 summarize the degradation temperatures, maximum decomposition temperature (T_{max}), and char yield of the prepared systems. The neat EEPO resin exhibited an initial degradation temperature (T_5%) of 140.2°C and reached 50% mass loss at 367.2°C, with a char residue of 4.0. The incorporation of nanofillers shifted the onset of degradation to higher temperatures, indicating enhanced thermal resistance. For the EEPO/GNP-MMT (0.5:0) sample, T_5% increased to 191.5°C and T_{10}% to 254.4°C, while T_{50}% remained nearly unchanged (371.2°C). The improvement in early-stage stability can be attributed to the inhibitory effect of

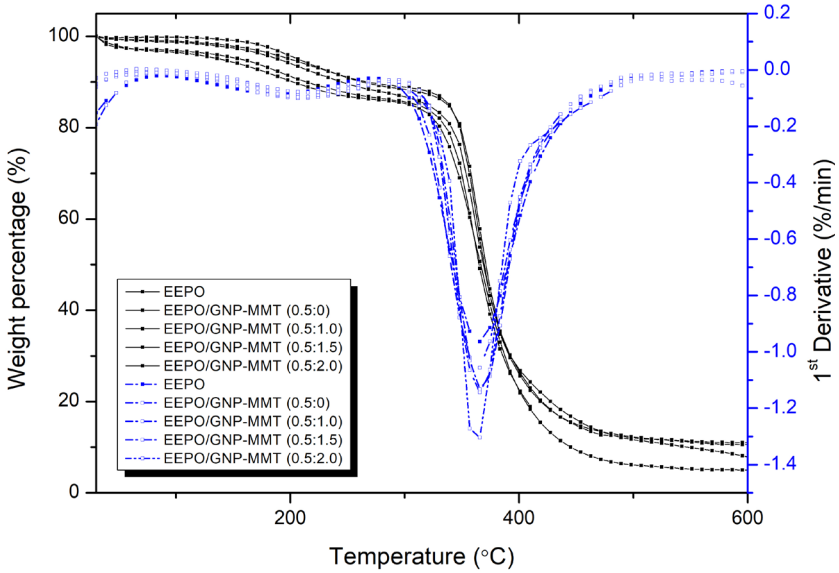

Fig. 5. TGA and DTG analysis of EEPO resin blends incorporated with the hybrid of GNP-MMT of different loadings and ratios

TABLE 2

Thermal properties of the EEPO resin blends containing the hybrid of GNP-MMT of different loadings and ratios

Formulation	Degradation Temperature (°C) at 5, 10, 50% weight loss			T_{max}	Char yield (%) at 700°C
	5%	10%	50%		
EEPO	140.2	203.2	367.2	369.5	4.0
EEPO/GNP-MMT (0.5:0)	191.5	254.4	371.2	367.6	6.1
EEPO/GNP-MMT (0.5:1.0)	200.6	264.9	371.2	366.5	9.2
EEPO/GNP-MMT (0.5:1.5)	208.9	261.3	372.4	367.2	10.3
EEPO/GNP-MMT (0.5:2.0)	190.5	246.0	370.9	366.0	5.1

GNP, which limits the diffusion of volatile degradation products and slows chain scission [21], [22]. The modest increase in char yield (6.1%) further supports the improvement in stability. The addition of 1.0 phr MMT in the EEPO/GNP-MMT (0.5:1.0) resulted in a more pronounced enhancement, with T_5% and T_{10}% increasing to 200.6°C and 264.9°C, respectively. The significant increase in char residue (9.2%) indicates that the hybrid filler promotes the formation of a thermally stable carbonaceous layer during decomposition. This finding is consistent with the synergistic interaction between well-dispersed GNPs and MMT platelets, which act as mass and heat transport barriers, thereby slowing the degradation process [23].The EEPO/GNP-MMT (0.5:1.5) formulation exhibited the best overall thermal performance, recording the highest T_5% (208.9°C) and a 10.3% increase in char yield. The slightly higher T_{50}% (372.4°C) compared with the other hybrids further supports the effectiveness of the dual-filler network at this ratio. The optimized dispersion of GNPs and MMT at this loading likely provides a three-dimensional barrier structure, reducing polymer chain mobility and suppressing thermal decomposition [23]. In contrast, at the highest MMT loading EEPO/GNP-MMT (0.5:2.0), T_5% and T_{10}% decreased to 190.5°C and 246.0°C, respectively, approaching the stability of the single-filler GNP system. The reduction in thermal stability is attributed to MMT agglomeration at high concentrations, which disrupts uniform filler dispersion and weakens the synergistic interaction with GNPs [5,24]. The lower char yield (5.1%) also supports this interpretation, suggesting that fewer effective barrier structures were formed to slow the degradation process. Overall, these results confirm that GNP-MMT hybrid incorporation significantly enhances the thermal stability of EEPO resins, with optimal performance achieved at the EEPO/GNP-MMT (0.5:1.5) ratio. However, at higher MMT contents, filler agglomeration and reduced interfacial compatibility outweigh the beneficial barrier and reinforcement effects.

3.3.2. Different Scanning Calorimetry (DSC)

Differential scanning calorimetry (DSC) was conducted to examine the thermal transitions of the EEPO/GNP-MMT hybrid systems, with particular emphasis on the glass transition temperature (T_g). As presented in Fig. 6, the neat EEPO system displayed a T_g of 55.54°C. Incorporation of 0.5 phr GNP without MMT (EEPO/GNP-MMT (0.5:0)) increased T_g to 58.54°C, indicating that even a small amount of graphene nanoplatelets can restrict chain mobility by creating interfacial interactions with the matrix. A more pronounced increase was observed when MMT was introduced. At 1.0 phr MMT (EEPO/GNP-MMT (0.5:1.0)), T_g rose significantly to 66.93°C, while a further increase to 1.5 phr MMT (EEPO/GNP-MMT (0.5:1.5))

yielded the highest T_g of 67.41°C. These increments suggest a strong synergistic interaction between GNP and MMT, where their co-dispersion produces a more homogeneous nanofiller network. This network forms a physical barrier to polymer chain relaxation and enhances interfacial adhesion, thereby requiring higher thermal energy for chain segment motion [23].

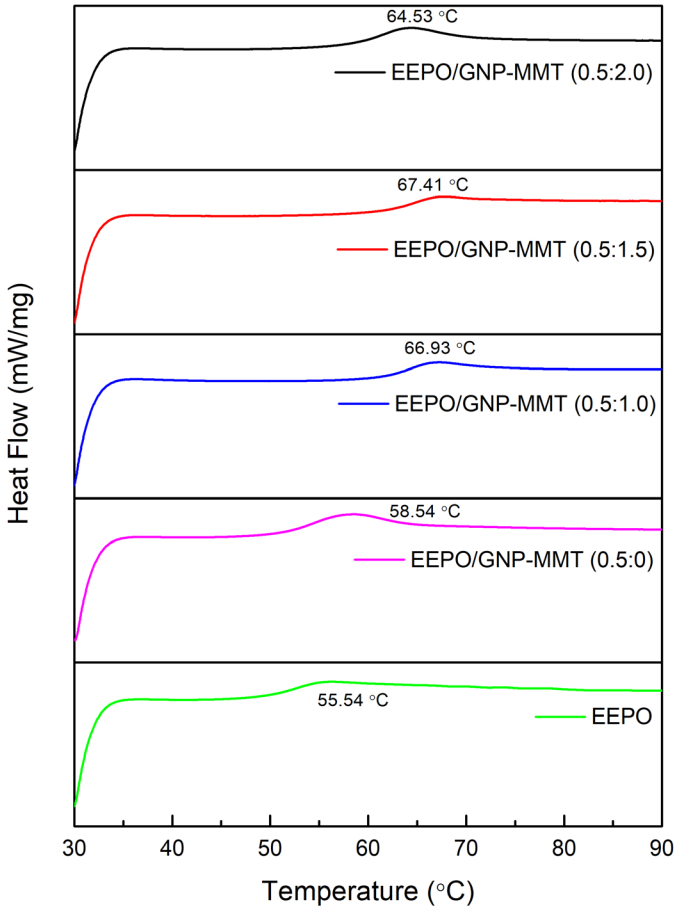

Fig. 6. DSC curves of the EEPO resin blends incorporated with the hybrid of GNP-MMT of different loadings and ratios

At higher MMT loading (EEPO/GNP-MMT (0.5:2.0)), T_g decreased slightly to 64.53°C. This reduction is attributed to nanofiller agglomeration, which disrupts the uniform distribution of GNP-MMT hybrids and creates interfacial voids between the nanofiller clusters and the surrounding matrix. These poorly bonded regions increase the free volume available for chain motion, thereby reducing the effectiveness of the restriction mechanism [20]. Consequently, while the addition of GNP and moderate amounts of MMT enhances chain rigidity and raises T_g, excessive MMT loading offsets the synergistic effect by promoting filler clustering, leading to lower thermal resistance [17], [20].

3.3.3. Scanning Electron Microscopy (SEM)

Fig. 7(a)-(d) shows the SEM micrographs of the fracture surfaces of EEPO resin-based nanocomposites reinforced with GNP-MMT hybrids at different ratios: GNP:MMT (phr): 0:0.5, 0.5:1.0, 0.5:1.5, and 0.5:2.0. Overall, the morphology of the fractured surfaces varies significantly with the MMT content, reflecting the differences in nanofiller dispersion, interfacial adhesion, and crack propagation mechanisms within the composites. For the EEPO/GNP-MMT (0.5:0) sample, a long and continuous crack path is observed, suggesting weaker filler-matrix adhesion and insufficient reinforcement, which allows cracks to propagate easily [25]. In the EEPO/GNP-MMT (0.5:1.0) sample, the cracks are still visible but less pronounced, indicating slightly improved dispersion and stronger interfacial interactions compared to the first composition. The EEPO/GNP-MMT (0.5:1.5) sample exhibits the most uniform and rough fracture surface, with no obvious long cracks. This morphology suggests better nanofiller distribution and stronger filler-matrix bonding, which forces the crack to propagate along a more tortuous path. A tortuous fracture surface generally requires higher energy for crack advancement, contributing to improved fracture resistance and superior mechanical properties [26]. In contrast, the EEPO/GNP-MMT (0.5:2.0) sample shows signs of non-uniform filler dispersion, where localized agglomeration likely occurred. These agglomerates can act as stress concentrators, leading to reduced interfacial adhesion and lower fracture resistance compared to the EEPO/GNP-MMT (0.5:1.5) formulation [27]. Overall, the SEM findings correlate well with the mechanical results, where the EEPO/GNP-MMT (0.5:1.5) composition exhibited the highest flexural strength and modulus. The improved performance at this formulation can be attributed to the combination of optimal filler dispersion, strong interfacial bonding, and greater resistance to crack initiation and propagation. Deviations from this ratio, either by increasing or decreasing MMT content, result in reduced mechanical performance due to poorer filler dispersion and weaker interfacial adhesion.

4. Conclusions

This work demonstrates that integrating GNP-MMT hybrid nanofillers into EEPO resin produces a pronounced synergistic effect, with the optimum balance achieved at a EEPO/GNP-MMT (0.5:1.5). At this composition, XRD analysis confirmed the greatest interlayer spacing of MMT, signifying improved dispersion, while SEM observations revealed a more uniform and rough fracture surface, reflecting stronger interfacial bonding. These microstructural features are consistent with the superior mechanical response, arising from efficient stress transfer and the minimization of stress concentration zones. Thermal analysis further verified the beneficial role of the hybrid system, as all filled samples exhibited higher stability compared with neat EEPO, with the EEPO/GNP-MMT (0.5:1.5) formulation showing the highest onset of degradation ($T_{5\%}$ = 208.9°C). The enhanced stability is linked to the cooperative action of GNP and MMT, where GNP assists in dispersing MMT platelets and jointly forms a protective barrier that delays polymer chain scission. However, excessive MMT incorporation (EEPO/GNP-MMT

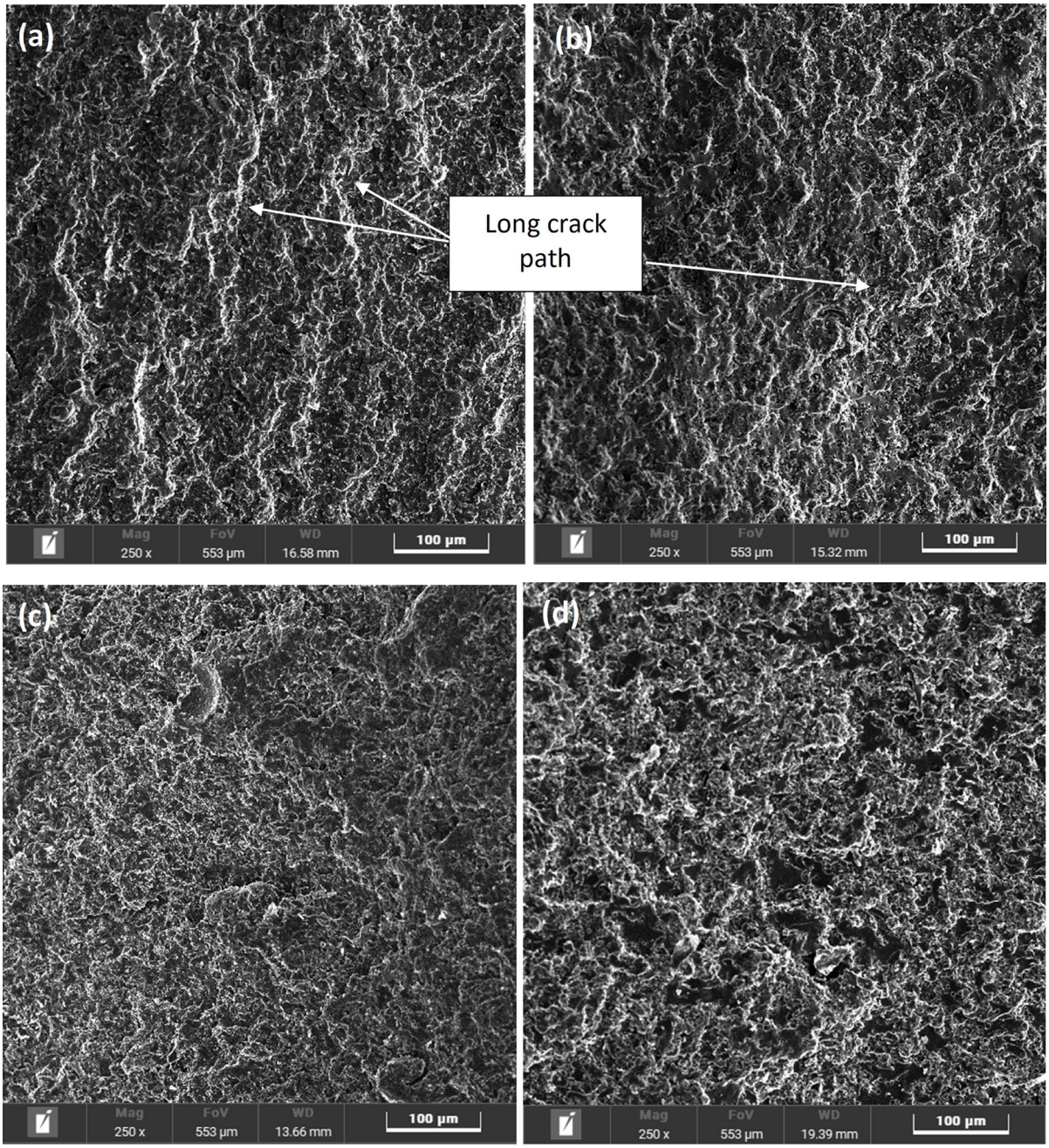

Fig. 7. SEM images of the EEPO resin blend containing GNP:MMT (a) 0.5:0; (b) 0.5:1.0; (c) 0.5:1.5; (d) 0.5:2.0

(0.5:2.0)) triggered filler agglomeration and, together with the intrinsic thermal conductivity of GNP, accelerated degradation instead of improving it. Collectively, these results highlight the effectiveness of GNP–MMT hybrids as a rational design pathway to tailor the mechanical and thermal properties of EEPO-based nanocomposites for advanced applications.

Acknowledgments

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