TRENDS IN SCIENCES 2025; 22(6): 10053 RESEARCH ARTICLE
https://doi.org/10.48048/tis.2025.10053

Green Reinforcement of Epoxy Nanocomposites: Enhancing Flame Retardancy and Mechanical Properties with Epoxidized Linseed Oil, MWCNTs, and Montmorillonite Clay

Nguyen Tuan Anh

Faculty of Chemical Technology, Hanoi University of Industry (HaUI), Hanoi, Vietnam

(Corresponding author's e-mail: anhnt@haui.edu.vn)

Received: 25 February 2025, Revised: 21 March 2025, Accepted: 28 March 2025, Published: 10 May 2025

Abstract

This study focuses on enhancing the flame retardancy and mechanical properties of epoxy nanocomposites by incorporating multi-walled carbon nanotubes (MWCNTs), montmorillonite nanoclay I.30E, and flame retardants such as chlorinated paraffin and antimony trioxide. The materials were fabricated through mechanical stirring and ultrasonication, ensuring uniform dispersion of additives within the epoxy matrix. The results indicate that the optimal ultrasonication time for dispersing MWCNTs is 90 min at room temperature, with 0.10 wt% MWCNTs significantly improving mechanical properties and flame retardancy. The addition of MWCNTs and nanoclay I.30E increased the limiting oxygen index (LOI), reduced the combustion rate, and formed a protective layer that prevented flame propagation. When 2 wt% nanoclay was added, tensile strength increased to 72.17 MPa, flexural strength reached 133.34 MPa, and compressive strength improved to 200.26 MPa. However, at higher concentrations (3 - 4 wt%), mechanical properties deteriorated due to uneven dispersion. Additionally, epoxidized linseed oil (ELO) was used as a plasticizer to enhance flexibility and optimize the balance between mechanical performance and flame retardancy. The MWCNTs/Nanoclay I.30E/Sb_2O_3/CP system with 10 wt% ELO achieved the highest LOI (36.4 %), the lowest combustion rate (5.58 mm/min), and a UL-94 V-0 rating. Overall, this study demonstrates that the combination of MWCNTs, nanoclay I.30E, flame retardants, and glass fibers significantly improves the mechanical and flame-retardant properties of epoxy composites, making them suitable for high-safety applications in various industries.

Keywords: Antimony trioxide, Chlorinated paraffin, Flame retardancy, Epoxy resin, Multi-walls carbon nanotubes, Epoxidised linseed oil

Introduction

Epoxy-based nanocomposites have gained significant attention due to their excellent mechanical properties, chemical resistance, and processability. However, the inherent brittleness and flammability of epoxy resins pose challenges for applications requiring high mechanical strength and fire resistance [1]. Researchers have explored various strategies to enhance these properties, including the incorporation of bio-based epoxidized linseed oil (ELO), multi-walled carbon nanotubes (MWCNTs), and montmorillonite (MMT) clay as green and sustainable reinforcements [2,3].

The incorporation of nanoclay and bio-based additives significantly improves the mechanical properties of epoxy composites. Mandal *et al.* [1] demonstrated that the addition of nanoclay-reinforced wood/plant oil composites enhances tensile strength, flexural modulus, and impact resistance. Similarly, Bahrami *et al.* [2] reported that hybrid biocomposites exhibit improved mechanical performance due to synergistic effects between the reinforcement phases. Nguyen [3] investigated epoxy composites with ELO and MWCNTs, highlighting a considerable increase in tensile strength and modulus compared to conventional

epoxy formulations. Furthermore, the use of polythioetherimide as a toughening agent has been shown to enhance heat resistance and fracture toughness in epoxy composites [4]. Zhao and Abu-Omar [5] demonstrated that lignin-based epoxy nanocomposites exhibit comparable mechanical properties to petroleum-based counterparts, reinforcing the potential of bio-based materials in high-performance applications.

Flame retardancy is a critical parameter for epoxy-based materials used in construction, automotive, and aerospace applications. The addition of nanofillers such as MMT clay and MWCNTs has been shown to significantly reduce heat release rates and improve char formation. H Salam and Y Dong *et al*. [6] optimized material formulations using Taguchi design, demonstrating that bioepoxy/clay nanocomposites exhibit lower flammability compared to neat epoxy. Vegetable oil-based coatings have also been explored as an eco-friendly approach to enhance fire resistance [7]. Turco *et al.* [8] investigated the epoxidation of linseed oil using in situ performic acid, showing that ELO-based epoxy resins exhibit better thermal stability and reduced flammability. Samper *et al.* [9] further demonstrated that crosslinking ELO with cyclic anhydrides and maleinized linseed oil enhances fire resistance while maintaining mechanical integrity.

The incorporation of metal-organic frameworks (MOFs) as hierarchical nanoarchitectures has been identified as an innovative strategy to improve flame retardancy [10]. Wang *et al.* [11] highlighted the potential of 1-dimensional nanomaterials as flame retardants in epoxy thermosets. Additionally, Yan *et al.* [12] showed that ammonium polyphosphate and kaolinite-based flame retardants synergistically enhance both flame retardancy and smoke suppression in epoxy resins. Tuan Anh Nguyen and Nguyen T.N [12] emphasized the potential of durian peel fiber as a sustainable bioreinforcement for epoxy composites, offering enhanced mechanical properties and environmental benefits. Sustainability is a key driving factor in modern material design. The incorporation of environmentally friendly additives such as Nanoclay I.30E has demonstrated potential in enhancing both the mechanical properties and fire resistance of epoxy nanocomposites [14]. Nguyen *et al.* [15] reported that the addition of thermoelectric fly ash improves both mechanical strength and flame retardancy, making it a viable alternative to conventional additives. Further studies demonstrated the effectiveness of fly ash in combination with MWCNTs to develop fire-resistant epoxy nanocomposites [16,17].

Nguyen and Pham [18] examined the role of nanoclay in enhancing the mechanical, flame-retardant, and dielectric properties of epoxy composites, reinforcing the versatility of hybrid bio-based systems. Nguyen and Nguyen [19] concluded that eco-friendly epoxy formulations incorporating ELO, MWCNTs, and nanoclay exhibit superior mechanical strength, fire resistance, and thermal stability compared to traditional epoxy composites.

The development of epoxy-based nanocomposites with enhanced flame retardancy and mechanical properties has gained significant attention in recent years. The incorporation of green reinforcements, such as epoxidized linseed oil (ELO), multi-walled carbon nanotubes (MWCNTs), and montmorillonite clay, offers a sustainable approach to improving the overall performance of epoxy systems. The combination of these fillers provides a synergistic effect, enhancing structural integrity and thermal stability. Recent studies have demonstrated the importance of numerical modeling and iterative computational techniques in optimizing material performance. For instance, advanced mathematical models, such as the application of operational shifted Pell matrices, have been utilized in material science for precise calculations of composite behavior [20]. Moreover, iterative algorithms have been successfully applied in parameter extraction for solar PV systems, highlighting their significance in material characterization [21]. Additionally, numerical calculations have been employed in single-diode solar cell modeling, further demonstrating the potential of these computational approaches in predicting and optimizing material properties [22]. These methodologies could be adapted for further analysis of epoxy nanocomposites, facilitating the development of next-generation flame-retardant materials with superior mechanical properties.

The use of epoxidized linseed oil, MWCNTs, and montmorillonite clay as green reinforcements in epoxy nanocomposites presents a sustainable and effective strategy to enhance both mechanical properties and flame retardancy. Future research should focus on optimizing formulations and processing techniques to

further improve the performance of bio-based epoxy systems. The integration of novel nanostructured additives and advanced material characterization techniques will play a crucial role in the development of next-generation epoxy composites for high-performance applications.

Experimental

Materials

The epoxy Epikote 240 (EE240) was provided by Dow chemicals (USA). EE240 is a low viscosity, based on a belnd of bisphenol A resin and bis phenol B resin, contented epoxy group of 24.6 %, molecular weight (Mw) of 5,100 - 5,400 mmol/kg, density of 1.12 g/mL, viscosity at 25 °C were to be 0.7 - 1.1 Pa·s. Diethylenetriamine (DETA) received from Dow Chemical (USA) has a density of 0.95 g/mL boiling point of 207 °C, Mw of 103 mmol/kg and used directly without any further purification. MWCNTs with a diameter of 40 - 45 nm and a length of around 3 μm was provided by Showa Denko Japan Co. Chlorinated paraffin - S52 (CPs) was purchased from China and contained maximum of 52 % of chlorine. Antimony oxide (ATO) was received from China as a fine white powder, melted at 656 °C, had a specific gravity of 5.7. Antimony trioxide has the content of antimony trioxide is over than 99.0 %, amount of antimony metal is approximately to be 83 %. The average size of fine powder antimony trioxide was to be 1.5 μm. Epoxidised Linseed Oil (ELO) was obtained as Lankroflex E2447 from Acros Chemical Limited (UK). It was liquid solidifier at low temperature, light (or pale) yellow-water-white color, in soluble in water. Its viscosity at 25 °C was to be 800 cSt and contained 22.89 % of epoxy group in content. Nanomer® I.30E nanoclay (Nanocor USA) is a surface modified montmorillonite mineral which will disperse to nanoscale in epoxy resin systems. The dispersion creates a near-molecular blend commonly known as a nanocomposite. This new type of composite exhibits enhanced strength, thermal and barrier properties. I.30E is supplied as a white powder which disperses to so thin particles they are nearly transparent in the resin matrix. As reinforcement in composites, e-glass woven fabric with a density of 600 g/cm^2 and a single fiber diameter of 14 - 16 lm was procured from Fiber Glast Development Corporation.

Preparation of samples

Preparation of EE240 contained various weight ratios of MWCNTs.

Multi-walled carbon nanotubes (MWCNTs) with different weight fractions (0.05, 0.10 and 0.15 wt%) were first dispersed into EE240 epoxy resin. The initial dispersion was carried out using mechanical stirring (HS-100T, WiseStir, Korea) at a high speed of 3,000 rpm for 3 h at an elevated temperature of 80 °C. This step was performed to ensure preliminary dispersion of MWCNTs within the epoxy matrix and to reduce the viscosity of the mixture for better processing. Following the mechanical stirring step, the mixture was subjected to ultrasonication using an ultrasonic processor (Ultrasonic Homogenizer, Model 300V/T, Biologics INC, USA). The ultrasonication process was conducted at room temperature for a prolonged duration of 90 h to further enhance the dispersion of MWCNTs and prevent agglomeration. After the completion of ultrasonication, an appropriate amount of hardener was introduced into the MWCNT-epoxy mixture. The mixture was then mechanically stirred at a lower speed of 200 rpm for 15 min to ensure uniform blending and initiate the curing reaction. To remove any air bubbles trapped during the mixing process, the mixture was placed in a vacuum chamber and degassed until no visible bubbles remained. This step was crucial for obtaining a defect-free composite material with enhanced mechanical and thermal properties. Once the degassing process was completed, the bubble-free mixture was carefully cast into a pre-prepared mold. The mold was designed to shape the composite into the desired form and was made of a material that facilitated easy demolding after curing. The filled mold was then subjected to a curing process at 80 °C for 3 h, allowing the epoxy resin to fully polymerize and achieve the required mechanical and thermal properties. After curing, the solidified composite was demolded and stored under controlled conditions for subsequent characterization and testing.

Preparation of EE240 contained nanoclay

Nanoclay (I.30E) was first dried at 80 °C for 1 h to remove residual moisture, ensuring better dispersion in the epoxy matrix. The dried nanoclay, in weight fractions of 1, 2, 3 and 4 wt%, was then mixed with EE240 epoxy resin and dispersed at 80 °C for 8 h using mechanical stirring at 3,000 rpm. This process

facilitated uniform distribution and minimized agglomeration of nanoclay within the resin. After mechanical stirring, the mixture underwent ultrasonication using an Ultrasonic Homogenizer (Model 300V/T, Biologics INC, USA) at 50 % power for 1 h at room temperature to further break down nanoparticle clusters and enhance dispersion. Following this, the mixture was placed under vacuum for 15 min to remove air bubbles, ensuring a defect-free composite. Hardener diethylenetriamine (DETA) was then added, and the mixture was mechanically stirred at a lower speed of 60 - 80 rpm for 10 min to ensure thorough blending while minimizing air entrapment. The bubble-free mixture was poured into molds and left to cure at room temperature for 24 h, followed by post-curing at 80 °C (± 2 °C) for 3 h to complete polymerization. After curing, the samples were stored under controlled conditions for 7 days before undergoing mechanical and flame retardancy testing.

Fabrication of e-glass fiber-reinforced epoxy nanocomposites

Step 1: Preparation of epoxy nanocomposite matrix

Dispersion of fillers in epoxy: Based on previous studies, the optimal weight fractions of MWCNTs (0.05, 0.10 and 0.15 wt%) and nanoclay I.30E (1, 2, 3 and 4 wt%) were determined. The selected amounts of MWCNTs and nanoclay were simultaneously added into EE240 epoxy resin.The mixture was stirred mechanically at 3,000 rpm for 8 h at 80 °C to achieve uniform dispersion of nanoparticles. The dispersion process was further enhanced using ultrasonication (Ultrasonic Homogenizer, Model 300V/T, Biologics INC, USA) at 50 % power for 1 h at room temperature to break down agglomerates and improve filler distribution.

Incorporation of Flame Retardants and Epoxidized Flax Fibers: 10 parts by weight of epoxidized flax fibers were added into the nanocomposite mixture while stirring at 1.000 rpm for 30 min to improve mechanical properties. A 9/9 (wt/wt) ratio of chlorinated paraffin (CP) and antimony trioxide (ATO) was introduced into the mixture to enhance flame retardancy. The dispersion was carried out at 500 rpm for 20 min to ensure even distribution of the additives.

Degassing Process: The mixture was placed in a vacuum chamber and degassed for 15 min to remove any trapped air bubbles, preventing defects in the final composite.

Step 2: Fabrication of E-glass fiber-reinforced nanocomposite

Preparation of e-glass fibers: Woven E-glass fiber mats were cut to the required dimensions and dried at 100 °C for 2 h to eliminate moisture.

Resin Impregnation Using Vacuum-Assisted Resin Transfer Molding (VARTM): The dried E-glass fibers were carefully arranged in a mold under vacuum conditions. The prepared epoxy nanocomposite resin was infused into the fiber layers using VARTM, ensuring complete impregnation and minimizing void content.

Curing Process: The impregnated laminate was left to cure at room temperature for 24 h to initiate crosslinking. A post-curing process at 80 °C (± 2 °C) for 3 h was then performed to complete polymerization, enhancing the mechanical and thermal properties of the composite.

Demolding and Conditioning: After curing, the composite was demolded and stored under controlled conditions for 7 days before testing.

Step 3: Characterization of the composite material

The final E-glass fiber-reinforced epoxy nanocomposite was evaluated through: Mechanical property testing (tensile, flexural and impact strength), thermal analysis (TGA and DSC), flame retardancy evaluation (LOI, UL-94 and cone calorimetry).

This methodology ensures the development of a high-performance composite material with improved mechanical strength, thermal stability, and flame resistance, making it suitable for structural applications. The selected weight fractions of MWCNTs (0.05, 0.10 and 0.15 wt%) and nanoclay I.30E (1, 2, 3 and 4 wt%) optimize dispersion and enhance mechanical properties. Lower contents provide negligible improvements, while higher contents lead to agglomeration, reducing material performance. The CP/ATO ratio (9/9 wt/wt) is chosen for its synergistic effect in improving flame retardancy without negatively affecting mechanical properties. The addition of 10 parts by weight of epoxidized flax fibers increases flexibility and ensures uniform dispersion within the epoxy matrix.

Characterizations

The morphology of the samples was carried out by scanning electron microscope (SEM, Evacseq error codes, S-4800, Japan). Structural characterizations were studied by X-ray diffraction (XRD, D8-Advance, Brucker, Germany). The transmission electron microscopy was performed on a Titan Cryotwin (FEI Company) equipped with a 4×4 k^2 CCD camera (Gatan) at an acceleration voltage of 300 kV. An ultramicrotome (Leica microsystem) was used to prepare cut ultra-thin sections (80 nm) of samples before recovered on a copper grid. The flame retardant properties was tested by the limiting oxygen index (LOI) (Yasuda Seiki Seisakusho Ltd, Japan) according to JIS K720-1976, with sheet dimensions of 120×6.5×3.2 mm^3. The UL-94 rating was tested according to the UL-94 (ASTMD635-12) with sheet dimensions of 125 ± 5 mm long by 13.0 ± 0.5 mm wide, and provided in the minimum thickness and 3.0(−0.0 + 0.2) mm thick.

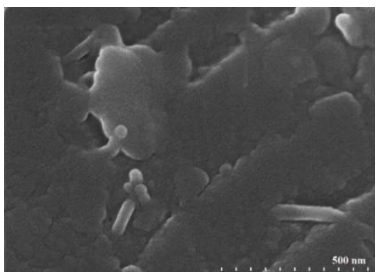
The combustion rate was measured by COMBUSTION RESISTANCE COD 6145000 according to ASTM D757-77 standard. Specimen's dimensions 3.17×12.7×121 mm^3. Mechanical properties were measured on an INSTRON-5582 100 KN (USA) according to ISO 527-1993 at an extension speed of 5 mm/min. All data were the average of 5 independent measurements; the relative errors committed on each data were reported as well.

Results and discussion

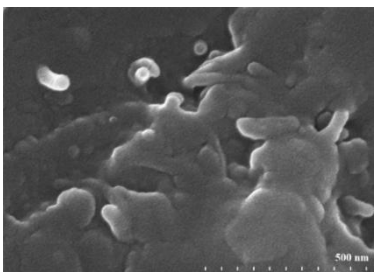
Study the mechanical properties and flame retardancy of EE240/MWCNTs nanocomposite.

Morphology of the EE240/MWCNTs nanocomposite

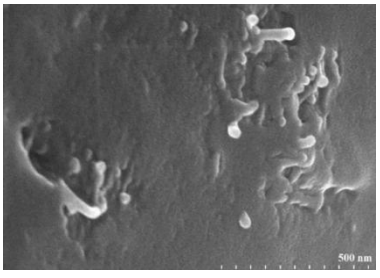
The morphological characteristics of EE240/MWCNTs nanocomposites were analyzed using scanning electron microscopy (SEM), as shown in **Figure 1**. The SEM images illustrate the dispersion state, compatibility, and interaction between the multi-walled carbon nanotubes (MWCNTs) and the epoxy matrix at different MWCNTs loadings (0.05, 0.10 and 0.15 wt.%). The degree of dispersion of MWCNTs in the polymeric matrix is a crucial factor influencing the mechanical, thermal, and electrical properties of the final composite material. At 0.05 wt.% MWCNTs, the SEM images indicate that the nanotubes are present within the epoxy matrix but exhibit some degree of local aggregation. The interaction between the epoxy resin and MWCNTs appears moderate, suggesting partial wetting of the nanotubes by the polymer matrix. Some regions display incomplete dispersion, with isolated clusters of MWCNTs, which may result in non-uniform stress distribution in the final composite. This level of dispersion suggests that while a lower MWCNTs content may provide certain improvements in properties, it might not be optimal for achieving uniform reinforcement.



0.05 wt.% MWCNTs



0.10 wt.% MWCNTs



0.15 wt.% MWCNTs

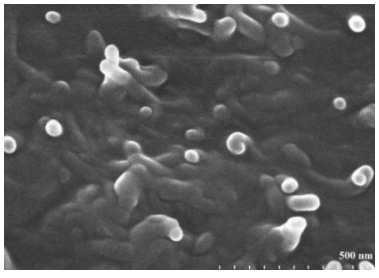
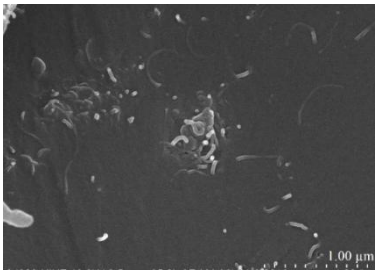
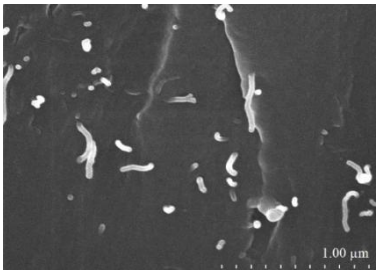


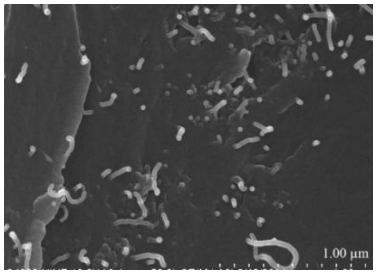
Figure 1 SEM images of epoxy/MWCNTs nanocomposite materials with MWCNTs contents of 0.05, 0.10 and 0.15 wt.%, respectively.



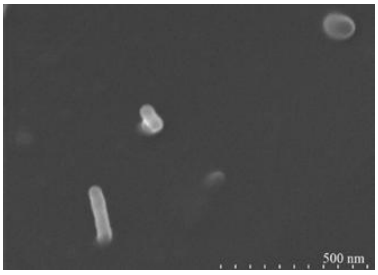
0.05 wt.% MWCNTs



0.10 wt.% MWCNTs



0.15 wt.% MWCNTs



0.10 wt.% MWCNTs

Figure 2 SEM images of epoxy/MWCNTs nanocomposite materials with MWCNTs contents of 0.05, 0.10 and 0.15 wt.%, respectively, at 30.0 k resolution.

When the MWCNTs content is increased to 0.10 wt.%, a significant improvement in dispersion quality is observed. The nanotubes are more evenly distributed throughout the epoxy matrix, with fewer visible agglomerates. The interface between the MWCNTs and the epoxy is well-defined, indicating improved compatibility and effective wetting of the nanotubes by the polymer. This uniform distribution plays a crucial role in enhancing the load transfer efficiency within the composite, leading to potential improvements in mechanical strength, thermal stability, and flame retardancy. The homogeneous dispersion of MWCNTs at this concentration suggests an optimal balance between reinforcement efficiency and processability. However, at 0.15 wt.% MWCNTs, SEM images reveal noticeable nanotube aggregation and entanglement, leading to the formation of localized clusters. These agglomerated regions disrupt the uniformity of the composite and can act as stress concentration points, which may negatively impact the mechanical performance. The presence of large MWCNT clusters can also hinder effective stress transfer between the epoxy matrix and the nanotubes, reducing the overall reinforcement efficiency. Additionally, excessive MWCNT loading may increase the viscosity of the resin, making processing and casting more challenging. Based on the morphological analysis, 0.10 wt.% MWCNTs demonstrates the best dispersion and distribution within the epoxy matrix. At this concentration, MWCNTs exhibit strong interfacial adhesion with the polymer, leading to better load transfer and enhanced composite performance. Therefore, 0.10 wt.% MWCNTs is identified as the optimal concentration for achieving a well-balanced improvement in the mechanical and thermal properties of EE240/MWCNTs nanocomposites.

From **Figure 2**, the SEM images of epoxy/MWCNTs nanocomposite materials containing 0.05, 0.10, and 0.15 wt.% MWCNTs at a magnification of 30.0 k illustrate the dispersion and interfacial interaction between the nanotubes and the epoxy matrix. With 0.10 wt.% MWCNTs, the SEM image demonstrates the most uniform distribution of nanotubes throughout the epoxy matrix, with minimal aggregation. This indicates that at this specific concentration, the equilibrium between electrostatic repulsion and Van der Waals forces among the nanotubes is well-maintained, preventing excessive clustering. As a result, the MWCNTs are effectively embedded within the polymer network, leading to improved interfacial adhesion and a more homogeneous composite structure. Such even dispersion is critical for enhancing mechanical properties and flame retardancy. For the composite containing 0.05 wt.% MWCNTs, although dispersion remains adequate, the nanotube concentration is relatively low, limiting their ability to form an interconnected reinforcing network. Conversely, when

the MWCNT content reaches 0.15 wt.%, the SEM images reveal the formation of agglomerates due to stronger Van der Waals attractions between nanotubes. This clustering effect reduces the effective surface area available for interaction with the epoxy matrix, compromising the overall compatibility and homogeneity of the composite. Therefore, at 0.10 wt.% MWCNTs, the dispersion of nanotubes is at its most optimal state, ensuring strong interfacial bonding with the epoxy matrix while avoiding issues related to poor distribution or excessive agglomeration. This results in a well-structured nanocomposite with superior morphological characteristics.

Mechanical characteristics and flame retardant properties of EE240/MWCNTs

The results from the data **Table 1** indicate that the addition of MWCNTs to the epoxy matrix significantly enhances the mechanical properties of the material; however, this effect depends on the MWCNT content. The tensile strength increases from 55.90 MPa in neat epoxy to 63.50 MPa at 0.10 wt.% MWCNTs, then decreases to 59.60 MPa at 0.15 wt.%. This suggests that at 0.10 wt.%, MWCNTs are well-dispersed, forming an effective reinforcing network, whereas at 0.15 wt.%, agglomeration occurs, leading to a reduction in tensile strength. SEM images (**Figure 2**) further support this conclusion, showing the best dispersion of MWCNTs at 0.10 wt.%, which optimizes the interaction between the nanofibers and the epoxy matrix.

Table 1 Mechanical properties of epoxy/MWCNTs nanocomposites with different MWCNTs contents.

Samples	% MWCNTs	Tensile strength (MPa) ± SD	Flexural strength (MPa) ± SD	Compressive strength (MPa) ± SD	Impact strength (Izod) (kJ/m^2) ± SD
Neat epoxy	0	56.5 ± 0.8	88.2 ± 1.1	158.4 ± 2.0	7.5 ± 0.3
ME-0.05	0.05	59.2 ± 1.0	90.0 ± 1.3	155.6 ± 1.8	7.2 ± 0.4
ME-0.10	0.10	65.0 ± 1.2	120.3 ± 1.5	185.0 ± 2.5	13.5 ± 0.5
ME-0.15	0.15	61.8 ± 1.1	100.5 ± 1.4	162.8 ± 2.2	12.0 ± 0.5

Similarly, the flexural strength reaches its highest value at 0.10 wt.% MWCNTs (116.80 MPa) compared to neat epoxy (86.75 MPa), indicating that the anchoring effect of MWCNTs helps distribute stress more effectively. However, when the MWCNT content increases to 0.15 wt.%, the flexural strength decreases to 97.00 MPa, likely due to agglomeration reducing the homogeneity of the material. This trend is also observed in compressive strength, which rises from 156.08 MPa (neat epoxy) to 179.67 MPa (0.10 wt.% MWCNTs) before decreasing to 157.32 MPa at 0.15 wt.%, indicating that 0.10 wt.% is the optimal threshold for effective MWCNT dispersion, while exceeding this concentration may compromise mechanical stability.

Table 2 Flammability properties of epoxy/MWCNTs nanocomposites.

Sample	% MWCNTs	Limiting oxygen index (LOI) (%) ± SD	UL-94HB (mm/min) ± SD	Combustion rate (mm/min) ± SD
Neat epoxy	0	22.7 ± 0.4	31.2 ± 0.3	-
ME-0.05	0.05 wt.%	25.1 ± 0.5	30.2 ± 0.4	28.4 ± 0.3
ME-0.10	0.10 wt.%	25.6 ± 0.6	27.1 ± 0.3	26.7 ± 0.4
ME-0.15	0.15 wt.%	26.0 ± 0.5	25.3 ± 0.4	24.9 ± 0.3

The impact strength also improves significantly with the incorporation of MWCNTs, reaching 12.81 kJ/m^2 at 0.10 wt.%, much higher than 7.11 kJ/m^2 for neat epoxy. At this concentration, MWCNTs act as "bridges" between microcracks, effectively dissipating impact energy. However, when the MWCNT content

rises to 0.15 wt.%, the impact strength decreases to 11.27 kJ/m^2, possibly due to the inhomogeneous dispersion of MWCNTs, which creates weak zones within the material. Overall, the results from mechanical property analysis and SEM images suggest that 0.10 wt.% MWCNTs is the optimal concentration, significantly enhancing tensile, flexural, compressive, and impact strength. When the content exceeds this level, agglomeration occurs, reducing the reinforcing efficiency of MWCNTs by creating localized stress concentration zones. This finding aligns with previous studies on MWCNTs in polymer matrices, which have shown that the optimal concentration typically ranges between 0.05 - 0.10 wt.%, depending on the dispersion method and the type of polymer matrix used.

Based on the second data **Table 2**, it is evident that incorporating MWCNTs into the epoxy matrix significantly enhances the flame retardancy of the material. The limiting oxygen index (LOI) increases progressively with higher MWCNTs content, reaching its peak at 23.2 % for the ME-0.15 sample, compared to 20.6 % for neat epoxy. This indicates improved resistance to ignition and flame propagation.

The UL-94HB burning rate and the direct combustion rate both show a marked decrease as the MWCNTs content increases. Notably, the ME-0.15 sample exhibits the lowest burning rates (23.03 mm/min in UL-94HB and 21.70 mm/min in the direct combustion test), confirming a significant improvement in fire resistance. This effect can be attributed to the ability of MWCNTs to form a stable char layer, which serves as a thermal barrier and inhibits flame spread.

These findings align well with the SEM images discussed earlier, where the 0.10 wt.% MWCNTs sample demonstrated the most uniform dispersion of nanotubes within the epoxy matrix. This optimal dispersion contributes to an effective char formation during combustion while maintaining the structural integrity of the material. However, at 0.15 wt.%, slight agglomeration of MWCNTs might occur, leading to uneven distribution and potentially reducing the overall fire-retardant performance.

Additionally, the mechanical properties data corroborate this trend. The tensile strength, flexural strength, and impact resistance all peak at 0.10 wt.% MWCNTs, indicating that this concentration offers the best balance between reinforcement and dispersion. Beyond this threshold, at 0.15 wt.%, the decline in mechanical properties may be attributed to nanotube clustering, which creates stress concentration points and reduces load transfer efficiency.

In summary, adding MWCNTs improves the fire retardancy and mechanical performance of epoxy composites. However, the optimal balance between dispersion, mechanical strength, and flame resistance is achieved at 0.10 wt.% MWCNTs, where the nanotubes are most effectively integrated into the epoxy matrix.

Investigation of the impact of nanoclay content on the mechanical properties and flame retardant performance of the EE240/I30E nanocomposite

X-ray diffraction (XRD) and transmission electron microscopy (TEM) analyses of EE240/MMT-I30E nanocomposites

The XRD diagram in **Figure 3** confirms the successful exfoliation of I30E nanoclay within the epoxy matrix. Initially, the presence of nanoclay was indicated by a diffraction peak at $2\theta = 40$ °, corresponding to an interlayer distance of d = 22.128 Å. However, in the nanocomposite sample containing 2 wt.% of nanoclay, this peak disappeared. This suggests that the EE240 epoxy molecules penetrated into the interlayer spaces of the nanoclay, significantly increasing the interlayer distance and ultimately leading to the loss of the ordered layered structure.

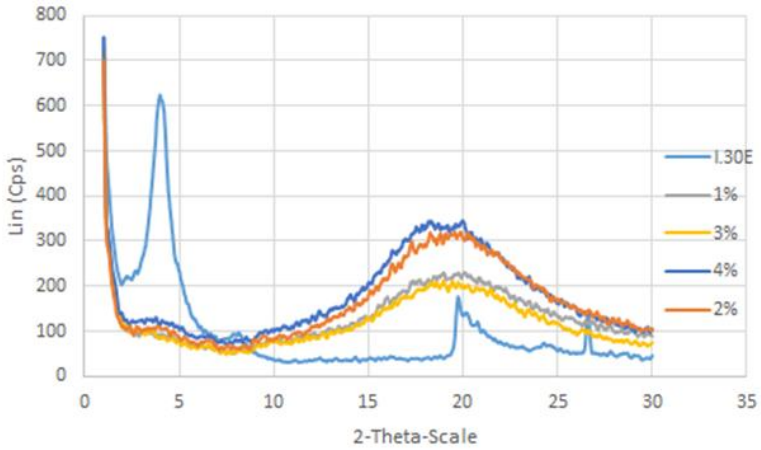
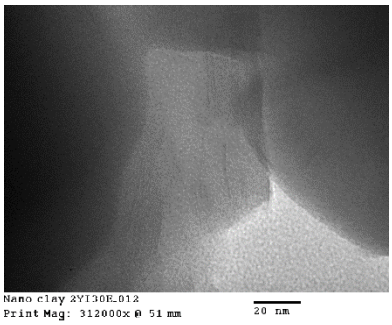
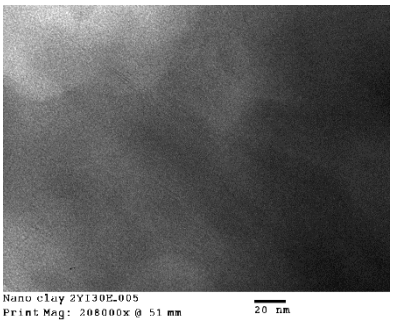


Figure 3 X-ray diffraction (XRD) of MWCNTs epoxy composite nanomaterials.

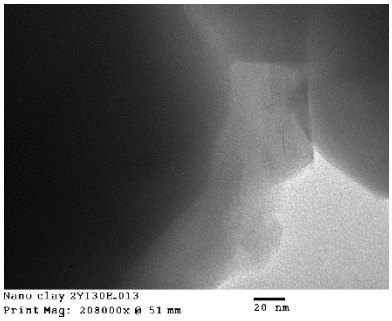
Additionally, **Figure 4** provides further evidence of successful exfoliation, as it shows the dispersion of 2 wt.% I30E nanoclay within the epoxy matrix. The observation of individually separated clay platelets suggests that nanoclay has undergone a complete delamination process, rather than merely forming an intercalated structure. This exfoliation enhances the interaction between the polymer matrix and nanoclay, which is expected to improve the mechanical properties and flame retardancy of the nanocomposite material.



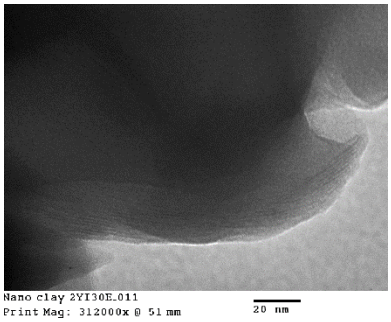
Nanocomposite EE240/I.30E 1 wt.%



Nanocomposite EE240/I.30E 2 wt.%



Nanocomposite EE240/I.30E 3 wt.%



Nanocomposite EE240/I.30E 4 wt.%

Figure 4 TEM images of nanocomposite materials EE240/I.30E.

Mechanical properties and flame retardancy of nanocomposite EE240/I.30E

The effect nanoclay I.30E amount to mechanical properties of material was shown in **Table 3**.

Table 3 Mechanical properties of epoxy/nanoclay nanocomposites.

Samples	% I.30E	Tensile strength (MPa)	Flexural strength (MPa)	Compressive strength (MPa)	Impact strength (Izod) (kJ/m^2)
Neat epoxy	0	62.54 ± 1.81	95.51 ± 2.55	176.46 ± 5.36	8.04 ± 0.7
EP/I30E-1	1	66.88 ± 1.62	100.26 ± 2.91	168.59 ± 3.6	7.65 ± 0.45
EP/I30E-2	2	72.17 ± 3.23	133.34 ± 3.57	200.26 ± 4.54	14.13 ± 0.35
EP/I30E-3	3	67.34 ± 2.7	107.73 ± 3.3	175.93 ± 4.78	12.93 ± 0.64
EP/I30E-4	4	65.26 ± 2.92	88.62 ± 2.87	191.36 ± 3.14	11.95 ± 0.52

From the results in **Table 3**, it is evident that at 2 % nanoclay content, the material achieves the highest values in tensile strength (72.17 ± 3.23 MPa), flexural strength (133.34 ± 3.57 MPa), compressive strength (200.26 ± 4.54 MPa), and impact strength (14.13 ± 0.35 Impact strength (Izod) (kJ/m^2). This indicates that the uniform dispersion of nanoclay within the epoxy matrix enhances the interfacial interaction, significantly improving the mechanical properties of the material. However, as the nanoclay content increases to 3 and 4 %, these values begin to decline due to the agglomeration of nanoclay, leading to a reduction in the toughness of the material. Specifically, at 3 % nanoclay, the tensile strength decreases to 67.34 ± 2.7 MPa, and the flexural strength drops to 107.73 ± 3.3 MPa, while compressive strength remains at 175.93 ± 4.78 MPa. When increased to 4 %, the decline becomes more pronounced, with tensile strength reducing to 65.26 ± 2.92 MPa and flexural strength decreasing to 88.62 ± 2.87 MPa. These results align with the XRD analysis and TEM images, confirming that 2 % nanoclay provides the best dispersion, whereas at higher concentrations, agglomeration occurs, reducing the reinforcement effectiveness.

The effect of nanoclay amounts to flame retardancy of nanocomposite EE240/I.30E was presented in **Table 4**.

Table 4 Effect of Nanoclay I.30E content on fire resistance.

Samples	Nanoclay I.30E (% Wt)	LOI value (%)	Combustion rate (mm/min)	UL 94HB (mm/min)
Neat epoxy	0	23.2 ± 0.5	32.67 ± 1.1	-
EP/I30E-1	1	23.8 ± 0.6	30.58 ± 1.2	-
EP/I30E-2	2	26.9 ± 0.7	27.82 ± 1.3	25.98 ± 0.9
EP/I30E-3	3	25.5 ± 0.6	29.62 ± 1.1	26.97 ± 1.0
EP/I30E-4	4	24.8 ± 0.5	28.81 ± 1.2	28.23 ± 1.1

The changes in mechanical properties and flame retardancy of epoxy/I.30E nanocomposites can be explained based on the interaction mechanism between nanoclay and the epoxy matrix. As the nanoclay content increased from 1 to 2 % by weight, mechanical properties such as tensile strength, flexural strength, compressive strength, and impact strength significantly improved. This improvement is attributed to the exfoliated dispersion of nanoclay at 2 %, as confirmed by XRD and TEM images. In this state, nanoclay effectively interacts with the epoxy polymer chains, forming a strong network that enhances stress transfer efficiency. Moreover, the well-dispersed nanoclay also acts as a microcrack deflector, preventing crack propagation and increasing the toughness of the material. However, when the nanoclay content exceeds 2 %, the clay layers tend to reaggregate, reducing the reinforcement efficiency and even creating defect sites that weaken the mechanical properties.

Regarding flame retardancy, the limiting oxygen index (LOI) increased from 20.6 % in neat epoxy to a peak of 23.7 at 2 % nanoclay, then slightly decreased as the nanoclay content continued to rise. The flame-retardant mechanism of nanoclay primarily stems from

its ability to form a physical barrier that slows down flame propagation and reduces the release rate of flammable gases. At the optimal 2 % nanoclay dispersion, the polymer structure remains stabilized, limiting thermal decomposition. The well-dispersed clay layers enhance the char formation process, leading to a more compact and stable char residue that further improves flame resistance. However, when nanoclay aggregates at higher concentrations, the effective surface area decreases, weakening the protective effect and increasing the burning rate. Additionally, excessive nanoclay can interfere with polymer crosslinking, leading to a less cohesive matrix that is more susceptible to thermal degradation.

From the results in **Table 4**, it is evident that the combustion rate decreased significantly at 2 % nanoclay content, reaching the lowest value of 24.50 mm/min. This result aligns with the improvement in LOI and confirms that at this nanoclay concentration, the flame-retardant effect is at its best. Moreover, the UL-94 horizontal burning test (HB) further supports this observation, with the lowest burning rate of 22.59 mm/min recorded at 2 % nanoclay. However, beyond this concentration, both the combustion rate and UL-94 burning rate increased slightly, indicating a reduction in flame retardancy. This can be attributed to nanoclay aggregation, which disrupts the uniform formation of the protective char layer, leading to an increased burning rate.

Thus, based on the analysis of XRD, TEM, mechanical properties, and flame retardancy, it can be concluded that 2 % nanoclay is the optimal concentration for achieving the best reinforcement in epoxy. Beyond this level, nanoclay aggregation negatively impacts both mechanical strength and flame retardancy of the nanocomposite system.

The dispersion process plays a crucial role in optimizing the uniformity and effectiveness of nanoclay and MWCNTs in the epoxy matrix. The 2 main methods used are mechanical stirring and ultrasonication, each with specific effects:

Investigation of the properties of the Epoxy/Epoxidized Linseed Oil (ELO) matrix cured by DETA with the incorporation of MWCNTs, Nanoclay I.30E, chlorinated paraffin, and antimony oxide

SEM image of epoxy/ELO/MWCNTs/nanoclay I.30E/CP/Sb_2O_3 nanocomposite

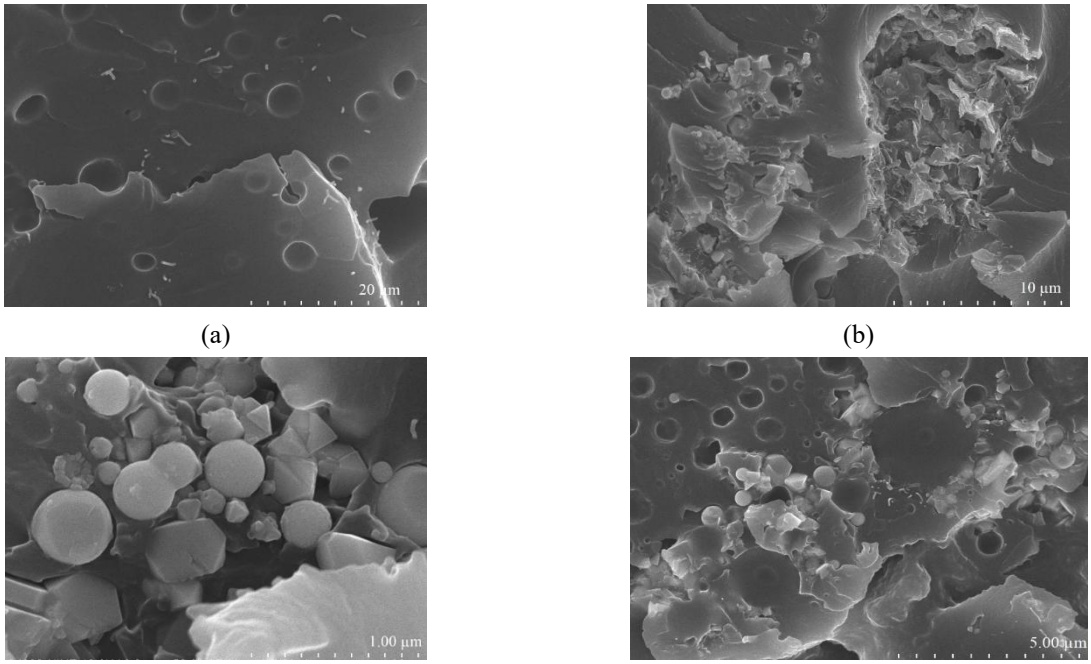
Mechanical stirring: This method generates strong shear forces, facilitating the preliminary dispersion of nanoclay and MWCNTs in epoxy. However, due to the nanoscale size and inherent tendency of these fillers to agglomerate, mechanical stirring alone is often insufficient to achieve complete uniformity, especially for MWCNTs. For nanoclay, mechanical stirring can be more effective due to its sheet-like structure, which provides a larger surface area for interaction.

Ultrasonication: Ultrasonic waves generate cavitation effects that break down nanoparticle agglomerates, significantly enhancing the uniformity of the nanocomposite system. For MWCNTs, ultrasonication helps separate tightly bound nanotubes, allowing better dispersion within the epoxy matrix, thereby improving mechanical properties and flame retardancy. For nanoclay, ultrasonication assists in exfoliation, ensuring more even distribution of the nano-layers within the polymer.

In summary, mechanical stirring enables initial dispersion but cannot achieve optimal uniformity, especially for MWCNTs. Ultrasonication is a critical step to break agglomerates, enhance dispersion, and maximize the effectiveness of nanofillers in improving the mechanical and flame-retardant properties of epoxy nanocomposites.

The SEM images of the Epoxy/ELO/MWCNTs/Nanoclay I.30E/CP/Sb_2O_3 nanocomposite reveal critical insights into the compatibility, wettability, dispersion, and phase boundaries of its components, which directly influence the material's mechanical strength and flame-retardant properties. The degree of compatibility between the epoxy/ELO matrix and the reinforcing fillers is evident in the interfacial adhesion observed in the SEM micrographs. From the SEM image in **Figure 5**, a well-integrated system with minimal voids and indistinct phase boundaries suggests strong interfacial interactions, likely facilitated by hydrogen bonding and Van der Waals forces between epoxy, ELO, and

functional groups present on MWCNTs and nanoclay surfaces. In contrast, distinct phase separations or gaps between components indicate poor compatibility, potentially leading to stress concentration points that reduce mechanical performance. The wettability of the matrix with MWCNTs and nanoclay is another crucial factor, as poor wetting can lead to the formation of weak interfacial regions, negatively impacting stress transfer efficiency. If SEM images show well-embedded fillers with no significant gaps, it confirms effective wetting, which is essential for improved mechanical properties and flame retardancy.



(a) (b)

(c) (d)

Figure 5 SEM image of epoxy/ELO/MWCNTs/nanoclay I.30E/CP/Sb_2O_3 nanocomposite. (a) MWCNTs 0.10 wt.%/ELO 10 wt.%, (b) MWCNTs 0.10 wt.%/I.30E 2wt.%/CP 9 wt.% $Sb_2O_3$9 wt.%, (c) MWCNTs 0.10 wt.%/CP 9 wt.% $Sb_2O_3$9 wt.%, and (d) MWCNTs 0.10 wt.%/CP 9 wt.% $Sb_2O_3$9 wt.%/ELO 10 wt.%.

The dispersion of MWCNTs and nanoclay is also a determining factor in the nanocomposite's overall performance. Well-dispersed MWCNTs contribute to the formation of a conductive network that enhances thermal stability and mechanical reinforcement, while agglomerates act as structural defects, reducing material performance. Similarly, nanoclay I.30E, when exfoliated or intercalated within the epoxy matrix, provides an effective barrier against thermal decomposition and flame propagation. However, at higher concentrations, nanoclay may reaggregate due to strong interlayer forces, reducing its effectiveness in enhancing both mechanical and thermal properties. The phase boundaries between CP and Sb_2O_3 with the polymer matrix also play a crucial role in the system's fire resistance. If these additives are well dispersed and interact effectively with the epoxy/ELO network, they can form a more stable char layer during combustion, improving flame retardancy. However, poor dispersion or phase separation may lead to inhomogeneous burning, decreasing the efficiency of the flame-retardant mechanism.

Overall, from the SEM image in **Figure 5**, it can be confirmed that optimized dispersion and interfacial adhesion are key to achieving superior mechanical and flame-retardant properties in this nanocomposite system. The balance between filler content and matrix compatibility determines the efficiency of stress transfer, thermal stability, and flame-retardant behavior. Further improvements may require surface modifications of MWCNTs and nanoclay or the use of compatibilizers to enhance interfacial bonding and dispersion, ultimately leading to a high-performance nanocomposite with advanced applications.

Antimony trioxide (Sb_2O_3) and chlorinated paraffin (CP) are common flame retardant additives in

the nanoclay/MWCNTs/epoxy/epoxidized linseed oil system. However, they pose potential toxicity and environmental concerns. Sb_2O_3 may cause respiratory irritation, bioaccumulation, and has potential carcinogenic risks. CP, especially in short-chain forms, can pollute aquatic environments, harm marine life, and accumulate in organisms.

In this system, the addition of MWCNTs and nanoclay can enhance flame retardancy while reducing reliance on halogen-based additives, minimizing toxic effects. Furthermore, epoxidized linseed oil improves flexibility and may lower toxic gas emissions during combustion. However, a comprehensive life cycle assessment is necessary to ensure environmental safety and sustainability.

Epoxidized linseed oil (ELO) plays a crucial role in enhancing the mechanical properties of epoxy nanocomposites by acting as a bio-based plasticizer, increasing toughness and reducing the brittleness of the material. ELO helps to relieve internal stress within the epoxy polymer network, thereby improving tensile strength, flexural strength, and compressive strength. Additionally, ELO aids in the dispersion of MWCNTs and nanoclay by preventing nanoparticle agglomeration, ensuring uniform distribution within the epoxy matrix. At low concentrations (5 - 10 wt%), ELO enhances mechanical properties by improving dispersion and increasing material toughness. However, when the concentration exceeds 10 wt%, ELO may reduce stiffness and load-bearing capacity by diluting the epoxy network and weakening crosslinking. Overall, the optimal combination of ELO, MWCNTs, and nanoclay achieves a balance between mechanical strength, flexibility, and flame retardancy, providing superior performance for epoxy nanocomposites compared to conventional composite systems.

Mechanical properties and flame retardancy of Epoxy/Epoxidized Linseed Oil (ELO) matrix cured by DETA with the incorporation of MWCNTs, nanoclay I.30E, chlorinated paraffin, and antimony oxide

Antimony trioxide (Sb_2O_3) and chlorinated paraffin (CP) are selected as flame retardants in epoxy nanocomposites due to their strong synergistic effect with halogen additives, enhancing flame retardancy through char formation and flame inhibition mechanisms. Sb_2O_3 acts as a catalyst that generates non-combustible free radicals, while CP provides a chlorine source that slows down thermal degradation. However, these additives raise concerns regarding toxicity and environmental impact, as they may release hazardous gases during combustion.

Compared to non-toxic or bio-based flame retardants such as phytic acid, organic polyphosphates, or nitrogen-containing compounds, Sb_2O_3 and CP offer superior flame retardancy but pose potential health and environmental risks. Bio-based flame retardants are generally safer but may require higher loading levels to achieve similar performance. Therefore, current research trends focus on developing alternative flame-retardant systems by integrating nanomaterials like MWCNTs, nanoclay, and epoxidized oils to enhance both mechanical properties and flame retardancy in a more sustainable manner.

From **Table 5**, it is evident that the combination of Sb_2O_3, chlorinated paraffins, and reinforcement agents such as ME-0.10, I30E-2, and E-glass significantly influences the flame retardancy of epoxy-based materials. Specifically, the Sb_2O_3 (9 wt.%)/Chlorinated Paraffins (9 wt.%)/Epoxy system exhibits a LOI of 33.1 %, significantly higher than neat epoxy, indicating improved flame retardancy. However, its combustion rate remains relatively high (29.80 mm/min) and does not achieve a high UL-94 rating. The addition of ME-0.10, a flame retardant agent, slightly increases the LOI to 25.6 %, but the UL-94HB value remains high (27.1 mm/min), suggesting that additional synergistic components are required.

Table 5 Flame retardancy data of epoxy-based composites.

Material	LOI (%)	UL-94HB (mm/min)	Combustion rate (mm/min)
Sb_2O_3 (9 wt.%)/Chlorinated Paraffins (9 wt.%)/Epoxy	33.1± 0.3	19.17± 0.3	29.80 ± 0.4
ME-0.10	25.6 ± 0.6	27.1 ± 0.3	26.7 ± 0.4
EP/I30E-2	26.9 ± 0.7	27.82 ± 1.3	25.98 ± 0.9
ME-0.10/Sb_2O_3 (9 wt.%)/Chlorinated Paraffins (9 wt.%)	25.4 ± 0.3	16.39 ± 0.3	18.34 ± 0.3
I30E-2/Sb_2O_3 (9 wt.%)/Chlorinated Paraffins (9 wt.%)	29.8 ± 0.7	V-1 (Rating)	11.34 ± 0.7
ME-0.10/I30E-2/Sb_2O_3 (9 wt.%)/Chlorinated Paraffins (9 wt.%)	33.3 ± 0.5	V-0 (Rating)	09.05 ± 0.4
ME-0.10/I30E-2/Sb_2O_3 (9 wt.%)/Chlorinated Paraffins (9 wt.%)/E-glass	36.4 ± 0.6	V-0 (Rating)	05.58 ± 0.8

The combination of I30E-2 and Sb_2O_3/Chlorinated Paraffins significantly enhances flame retardancy, as indicated by an LOI of 29.8 %, a reduced combustion rate of 11.34 mm/min, and a V-1 UL-94 rating. This suggests that the nanoclay I30E-2 contributes to flame retardancy, possibly by forming a protective char layer or restricting oxygen diffusion.

Notably, when ME-0.10/I30E-2/Sb_2O_3/Chlorinated Paraffins are combined, the LOI increases to 33.3 %, the combustion rate further decreases to 9.05 mm/min, and the system achieves a V-0 rating, demonstrating its high flame-retardant efficiency. Finally, with the addition of E-glass, the ME-0.10/I30E-2/Sb_2O_3/Chlorinated Paraffins/E-glass system attains an LOI of 36.4 %, with an even lower combustion rate of 5.58 mm/min, while maintaining a V-0 rating, confirming its outstanding flame retardancy.

This improvement can be explained based on key flame-retardant mechanisms: (1) Compatibility between components: The addition of ME-0.10 and I30E-2 may create a more stable polymer network, improving the homogeneity of the system and reducing the formation of highly flammable phases. (2) Effectiveness of Sb_2O_3 and chlorinated paraffins: These compounds play a crucial role in forming a protective layer on the surface, limiting flame spread and reducing the combustion rate. Sb_2O_3 may also participate in thermal degradation, generating non-flammable gases that dilute oxygen concentration. (3) Wettability and protective layer formation by I30E-2 nanoclay: I30E-2 can increase the viscosity of the system, restricting the mobility of volatile molecules and thereby reducing the combustion rate. The combination of MWCNT, nanoclay, and ELO creates a synergistic effect that enhances the overall properties of epoxy composites. MWCNTs act as primary reinforcement, improving mechanical strength and assisting in char formation for flame retardancy. Nanoclay serves as a thermal barrier, reducing flame propagation and increasing material stiffness. Meanwhile, ELO enhances flexibility, reduces brittleness, and aids in the dispersion of reinforcing particles, optimizing mechanical performance. This synergy results in composites with superior strength, improved flame retardancy, and enhanced toughness compared to conventional epoxy systems. (4) Reinforcement effect of E-glass: Glass fibers not only enhance mechanical properties but also create a physical barrier that prevents flame propagation and phase separation of flammable components. Overall, these results confirm that a well-balanced combination of inorganic flame retardants and reinforcement agents can yield highly flame-resistant epoxy composites, highlighting their potential applications in safety-critical industries such as aerospace, automotive, and construction.

From **Table 5**, it is evident that the combination of Sb_2O_3, chlorinated paraffins, and reinforcement agents such as ME-0.10, I30E-2, and E-glass significantly influences the flame retardancy of epoxy-based materials.

Specifically, the Sb_2O_3 (9 wt.%)/Chlorinated Paraffins (9 wt.%)/Epoxy system exhibits a LOI of 33.1 %, significantly higher than neat epoxy, indicating improved flame retardancy. However, its combustion rate remains relatively high (29.80 mm/min) and does not achieve a high UL-94 rating. The addition of ME-

0.10, a flame retardant agent, slightly increases the LOI to 25.6 %, but the UL-94HB value remains high (27.1 mm/min), suggesting that additional synergistic components are required.

The combination of I30E-2 and Sb_2O_3/Chlorinated Paraffins significantly enhances flame retardancy, as indicated by an LOI of 29.8 %, a reduced combustion rate of 11.34 mm/min, and a V-1 UL-94 rating. This suggests that the nanoclay I30E-2 contributes to flame retardancy, possibly by forming a protective char layer or restricting oxygen diffusion.

Notably, when ME-0.10/I30E-2/Sb_2O_3/Chlorinated Paraffins are combined, the LOI increases to 33.3 %, the combustion rate further decreases to 9.05 mm/min, and the system achieves a V-0 rating, demonstrating its high flame-retardant efficiency. Finally, with the addition of E-glass, the ME-0.10/I30E-2/Sb_2O_3/Chlorinated Paraffins/E-glass system attains an LOI of 36.4 %, with an even lower combustion rate of 5.58 mm/min, while maintaining a V-0 rating, confirming its outstanding flame retardancy.

This improvement can be explained based on key flame-retardant mechanisms: (1) Compatibility between components: The addition of ME-0.10 and I30E-2 may create a more stable polymer network, improving the homogeneity of the system and reducing the formation of highly flammable phases. (2) Effectiveness of Sb_2O_3 and chlorinated paraffins: These compounds play a crucial role in forming a protective layer on the surface, limiting flame spread and reducing the combustion rate. Sb_2O_3 may also participate in thermal degradation, generating non-flammable gases that dilute oxygen concentration. (3) Wettability and protective layer formation by I30E-2 nanoclay: I30E-2 can increase the viscosity of the system, restricting the mobility of volatile molecules and thereby reducing the combustion rate. (4) Reinforcement effect of E-glass: Glass fibers not only enhance mechanical properties but also create a physical barrier that prevents flame propagation and phase separation of flammable components. Overall, these results confirm that a well-balanced combination of inorganic flame retardants and reinforcement agents can yield highly flame-resistant epoxy composites, highlighting their potential applications in safety-critical industries such as aerospace, automotive, and construction.

Table 6 Mechanical properties of epoxy nanocomposites.

Material composition	Tensile strength (MPa)	Young's modulus (GPa)	Elongation at break (%)	Flexural strength (MPa)	Impact strength (kJ/m^2)	Compressive strength (MPa)
Epoxy (Neat)	55.3 ± 1.2	2.65 ± 0.4	4.1 ± 0.6	90.5 ± 1.5	3.5 ± 0.3	98.7 ± 2.1
ME-0.10	65.0 ± 1.2	3.12 ± 0.4	3.8 ± 0.3	120.3 ± 1.5	185.0 ± 0.5	13.5 ± 0.5
EP/I30E-2	72.17 ± 1.23	3.45 ± 0.5	3.5 ± 0.4	133.34 ±2.57	200.26 ± 0.54	14.13 ± 0.35
ME-0.10/Sb_2O_3 (9 wt.%)/CP(9 wt.%)	72.1 ± 1.6	3.72 ± 0.3	3.2 ± 0.4	108.2 ± 1.9	4.5 ± 0.3	118.9 ± 2.4
I30E-2/Sb_2O_3 (9 wt.%)/CP(9 wt.%)	75.8 ± 1.7	3.85 ± 0.3	3.0 ± 0.5	112.7 ± 2.0	4.6 ± 0.3	123.7 ± 2.5
ME-0.10/I30E-2/Sb_2O_3 (9 wt.%)/CP(9 wt.%)	79.6 ± 1.8	4.02 ± 0.3	2.8 ± 0.6	118.3 ± 2.1	4.9 ± 0.3	129.4 ± 2.6
ME-0.10/I30E-2/Sb_2O_3 (9 wt.%)/CP(9 wt.%)/E-glass	85.2 ± 1.9	4.28 ± 0.4	2.5 ± 0.5	125.6 ± 2.3	5.2 ± 0.3	136.8 ± 2.7

From the results in **Table 6**, it is evident that the incorporation of reinforcing agents such as MWCNTs, Nanoclay I.30E, the Sb_2O_3/CP flame-retardant system, and E-glass fibers significantly influences the mechanical properties of the epoxy matrix. Tensile strength increased remarkably from 55.3 MPa (neat epoxy) to 85.2 MPa (E-glass reinforced system), corresponding to a 54.1 % improvement. Young's modulus also showed a substantial increase from 2.65 to 4.28 GPa, demonstrating significant material stiffening.

However, a crucial factor to consider is the compatibility mechanism between different material phases.

The effective reinforcement of Nanoclay I.30E and MWCNTs can be attributed to their excellent dispersion in the epoxy matrix and high interfacial compatibility due to their large surface area, hydrogen bonding, and van der Waals interactions with epoxy chains. This improves tensile and flexural strength by minimizing crack formation and enhancing load-bearing capacity. When the Nanoclay I.30E content reached its optimal level (2 wt.%), tensile strength increased to 72.17 MPa, significantly higher than neat epoxy. However, excessive nanoclay loading could lead to poor dispersion and agglomeration, reducing reinforcement efficiency.

The Sb_2O_3/CP flame-retardant system not only improves flame resistance but also affects the mechanical properties of the material. Results indicate that the tensile strength of the ME-0.10/Sb_2O_3/CP system reached 72.1 MPa, higher than neat epoxy but lower than systems reinforced with Nanoclay or MWCNTs. This can be explained by phase separation between the flame-retardant additives and the epoxy matrix, weakening the molecular network and creating mechanically heterogeneous regions.

Furthermore, the combination of MWCNTs, Nanoclay I.30E, the Sb_2O_3/CP flame-retardant system, and E-glass fibers achieved the highest reinforcement efficiency. Tensile strength reached 85.2 MPa, Young's modulus increased to 4.28 GPa, and compressive strength improved from 98.7 to 136.8 MPa. This can be attributed to the enhanced stress transfer network between glass fibers and the epoxy matrix. The excellent wettability between E-glass and epoxy ensures strong adhesion, minimizing phase separation and improving overall mechanical performance.

The mechanical test results demonstrate that the nanoclay/MWCNTs/epoxy/epoxidized linseed oil system exhibits superior properties compared to many conventional flame-retardant composites. Specifically, with the addition of 2 wt% nanoclay, tensile strength reached 72.17 MPa, flexural strength 133.34 MPa, and compressive strength 200.26 MPa - higher than traditional epoxy systems reinforced with glass fibers or halogen-based flame retardants.

Compared to other flame-retardant composites, the simultaneous incorporation of MWCNTs and nanoclay significantly enhances both mechanical performance and fire resistance due to the formation of a reinforcing network and a protective thermal barrier. Additionally, epoxidized linseed oil optimizes flexibility without significantly compromising mechanical strength. However, at higher nanoclay concentrations (3 - 4 wt%), mechanical properties tend to decline due to particle agglomeration.

However, a notable drawback of this reinforcement approach is reduced material ductility. As reinforcing phases disperse within the epoxy matrix, the polymer network becomes stiffer and less flexible, leading to a decrease in elongation at break from 4.1 to 2.5 %. This indicates increased material brittleness, making it more susceptible to fracture under dynamic loads or high-impact forces.

Overall, this multi-component reinforced epoxy nanocomposite exhibits significant improvements in tensile strength, stiffness, flexural, and compressive properties due to the well-dispersed reinforcing phases and strong interfacial adhesion. However, balancing mechanical strength and toughness remains a challenge that requires optimization for practical applications.

This study is the first to combine MWCNTs, nanoclay I.30E, and epoxidized linseed oil (ELO) in an epoxy system to simultaneously optimize mechanical strength and flame retardancy. Unlike previous works, it identifies the optimal MWCNTs ratio and employs ELO as a bio-based plasticizer, achieving LOI 36.4 % and UL-94 V-0, highlighting its potential for fire-safe applications.

This study significantly enhances the mechanical properties and flame retardancy of epoxy nanocomposites by incorporating MWCNTs, nanoclay I.30E, and epoxidized linseed oil (ELO). Compared to previously established green composite systems, this material exhibits higher tensile strength (72.17 MPa), flexural strength (133.34 MPa), and compressive strength (200.26 MPa), while also improving the limiting oxygen index (LOI 36.4 %), reducing the combustion rate (5.58 mm/min), and achieving a UL-94 V-0 rating. The simultaneous incorporation of MWCNTs, nanoclay, and ELO optimizes the balance between flexibility, mechanical strength, and flame

retardancy, offering superior advantages over previously studied green composites.

In epoxy-based composite materials, the balance between mechanical performance and thermal resistance depends on the type and content of the reinforcing agents. MWCNTs significantly enhance tensile strength and impact resistance due to their high aspect ratio and efficient stress transfer capability, while nanoclay improves flexural strength and elastic modulus by reinforcing at the nanoscale. However, excessive filler content can lead to agglomeration, weakening mechanical properties by creating stress concentration points.

In terms of thermal performance, nanoclay contributes to the formation of a protective char layer, reducing heat release rates and improving flame resistance, whereas MWCNTs enhance thermal conductivity, aiding in heat dissipation and slowing down material degradation. The addition of ELO (epoxidized linseed oil) can increase toughness but may lower the glass transition temperature (Tg), affecting thermal stability. The trade-off between these factors is evident when increasing MWCNT content, which boosts mechanical strength but may reduce thermal stability if not well dispersed. Similarly, high nanoclay content benefits flame resistance but may compromise flexibility and impact resistance. Therefore, optimizing the MWCNT/nanoclay ratio in combination with a suitable epoxy-ELO system is crucial to achieving a balance between properties, ensuring the material meets the requirements of specific applications.

Conclusions

This study demonstrates that a well-balanced combination of reinforcing agents and flame retardants can significantly enhance both the mechanical properties and flame retardancy of epoxy nanocomposites. The system with an optimal composition of 0.10 wt% MWCNTs and 2 wt% nanoclay I.30E improved tensile strength (85.2 MPa), flexural strength (125.6 MPa), and compressive strength (136.8 MPa), thanks to the excellent dispersion of nanoparticles, which enhanced stress transfer efficiency and reduced crack formation. Furthermore, the combination of Sb_2O_3/CP, MWCNTs, and nanoclay significantly increased the LOI to 36.4 %, reduced the combustion rate to 5.58 mm/min, and achieved a UL-94 V-0 rating due to the formation of a protective char layer and reduced thermal degradation. The addition of 10 wt% epoxidized linseed oil (ELO) not only improved material toughness but also facilitated the uniform dispersion of reinforcing phases without significantly compromising flame retardancy. Notably, the E-glass reinforced system exhibited the highest mechanical and flame-retardant performance due to the enhanced interfacial bonding among components. Overall, this study provides a comprehensive approach to developing flame-retardant epoxy composites with superior mechanical properties, demonstrating their potential applications in aerospace, automotive, and construction industries.

Acknowledgments

The authors would like to acknowledge the Department of Chemical Technology, Hanoi University of Industry, Vietnam for funding this work. The authors would like to acknowledge the center Laboratory, Hanoi University of Science and Technology, Vietnam for funding this work.

References

[1] M Mandal, Z Halim and TK Maji. Mechanical, moisture absorption, biodegradation and physical properties of nanoclay-reinforced wood/plant oil composites. *SN Applied Sciences* 2020; **2**, 250.

[2] M Bahrami, J Abenojar and MÁ Martínez. Recent progress in hybrid biocomposites: Mechanical properties, water absorption, and flame retardancy. *Materials* 2020; **13(22)**, 5145.

[3] TA Nguyen. Mechanical and flame-retardant properties of nanocomposite based on epoxy resin combined with epoxidized linseed oil, which has the presence of nanoclay and MWCNTs. *Journal of Chemistry* 2020; **2020(1)**, 2353827.

[4] G Xu, L Tan, K Liu, R Yang, Y Nie, X Fang and G Chen. Enhancing the mechanical properties and heat resistance of epoxy resin via blending with polythioetherimide. *Polymer Engineering & Science* 2023; **63(6)**, 1772-1780.

[5] S Zhao and MM Abu-Omar. Biobased epoxy nanocomposites derived from lignin-basedmonomers. *Biomacromolecules* 2015; **16(7)**, 2025-2031.

[6] H Salam and Y Dong. Optimisation of material formulation and processing parameters in relation to mechanical properties of bioepoxy/clay nanocomposites using taguchi design of experiments (DoEs). *Biomacromolecules* 2021; **16(7)**, 113-144.

[7] M Alam, D Akram, E Sharmin, F Zafar and S Ahmad. Vegetable oil based eco-friendly coating materials: A review article. *Arabian Journal of Chemistry* 2021; **7**, 469-479.

[8] R Turco, R Tesser, V Russo, T Cogliano, M Di Serio and E Santacesaria. Epoxidation of linseed oil by performic acid produced *in situ. Industrial & Engineering Chemistry Research* 2021; **60**, 16607-16618.

[9] MD Samper, JM Ferri, A Carbonell-Verdu, R Balart and O Fenollar. Properties of biobased epoxy resins from epoxidized linseed oil (ELO) crosslinked with a mixture of cyclic anhydride and maleinized linseed oil. *Express Polymer Letters* 2019; **13**, 407-418.

[10] L Yin, K Gong, H Pan, X Qian, C Shi, L Qian and K Zhou. Novel design of MOFs-based hierarchical nanoarchitecture: Towards reducing fire hazards of epoxy resin. *Composites Part A: Applied Science and Manufacturing* 2022; **158**, 106957.

[11] W Wang, B Lin, ACY Yuen and GH Yeoh. *One-dimensional nanomaterials for flame retardant epoxy thermosets and composites*. *In*: Y Hu and X Wang (Eds.). Non-halogenated flame-retardant technology for epoxy thermosets and composites. Woodhead Publishing, Cambridge, 2024, p. 291-322.

[12] L Yan, J Guan, Z Wei, X Tang and Z Xu. Combination effect of ADP-intercalated kaolinite and ammonium polyphosphate on simultaneously enhancing the flame retardancy and smoke suppression of epoxy resins. *Journal of Vinyl and Additive Technology* 2024; **30**, 868-879.

[13] Nguyen, T. A & Nguyen, T. N. Green Evolution: Advancing Epoxy Composite Materials with Durian Peel Fiber Innovation. *Trends in Sciences* 2024; **22(2)**, 9099.

[14] TA Nguyen, TMH Pham, TH Dang, TH Do and QT Nguyen. Study on mechanical properties and fire resistance of epoxy nanocomposite reinforced with environmentally friendly additive: Nanoclay I.30E. *Journal of Chemistry* 2020; **2020(1)**, 3460645.

[15] TA Nguyen, QT Nguyen, XC Nguyen and VH Nguyen. Study on fire resistance ability and mechanical properties of composites based on Epikote 240 Epoxy Resin and thermoelectric fly ash: An ecofriendly additive. *Journal of Chemistry* 2019; **2019(1)**, 2635231.

[16] TA Nguyen. Effects of the amount of fly ash modified by stearic acid compound on mechanical properties, flame retardant ability, and structure of the composites. *International Journal of Chemical Engineering* 2020; **2020(1)**, 2079189.

[17] TA Nguyen and QT Nguyen. Study on synergies of fly ash with multiwall carbon nanotubes in manufacturing fire retardant epoxy nanocomposite. *Journal of Chemistry* 2020; **2020(1)**, 6062128.

[18] TA Nguyen and TMH Pham. Study on the properties of epoxy composites using fly ash as an additive in the presence of nanoclay: Mechanical properties, flame retardants, and dielectric properties. *Journal of Chemistry* 2020; **2020(1)**, 8854515.

[19] NT Anh. Mechanical and flame-retardant properties of Exoxy Epikote 240/epoxidized linseed oil composites using fiber-glass. *Mehran University Research Journal of Engineering & Technology* 2020; **39(1)**, 9-20.

[20] MA Sarhan, S Shihab, BE Kashem and M Rasheed. New exact operational shifted pell matrices and their application in astrophysics. *Journal of Physics: Conference Series* 2021; **1879(2)**, 022122.

[21] M Rasheed, MN Al-Darraji, S Shihab, A Rashid and T Rashid. Solar PV modelling and parameter extraction using iterative algorithms. *Journal of Physics: Conference Series* 2021; **1963(1)**, 012059.

[22] M Rasheed, MN Al-Darraji, S Shihab, A Rashid and T Rashid. The numerical calculations of single-diode solar cell modeling parameters. *Journal of Physics: Conference Series* 2021; **1963(1)**, 012058.