Original Article

Synergetic Effects of One and Two Dimension Nanofillers on the Mechanical Properties of Rubber Modified Epoxy Composites

A. Anand^{1*}, J. Dharanish¹, Bheemraj¹, D. C. Madaiah¹, H. Hema²

¹Department of Mechanical Engineering, The National Institute of Engineering, Mysuru, India. ²Department of Civil Engineering, The National Institute of Engineering, Mysuru, India.

**Corresponding Author : anand@nie.ac.in*

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Abstract - Graphene nanoplatelets (GnP), Multiwalled carbon nanotubes (M-cnT), and Halloysite nanotubes (HnT) exhibit a noteworthy synergistic impact in enhancing the mechanical properties of carboxyl-terminated butadiene-acrylonitrile-modified Epoxy (cr-Ep) nanocomposites. These nanofillers were incorporated into cr-Ep to form mono and hybrid nanocomposites. The introduction of 1-D M-cnT efficiently inhibits the stacking of individual 2-D GnP and HnT. A high contact area is created between the GnP/HnT with M-cnT and the cr-Ep matrix by the high strength and high aspect ratio of M-cnT, which can bridge neighboring GnP/HnT and prevent their aggregation. The mechanical properties of mono and hybrid nanocomposites were characterized in this study. The results showed that the mechanical properties of nanofiller reinforced cr-Ep composites were significantly improved. The tensile strength of GnP/cr-Ep and HnT/cr-Ep composites over the unfilled cr-Ep was only 24.5% and 20.5% higher, whereas the tensile strength of M-cnT/GnP/cr-Ep and M-cnT/HnT/cr-Ep composites was 39.5% and 27.6% greater than that of the cr-Ep alone. The M-cnT/cr-Ep and M-cnT/GnP/cr-Ep nanocomposites exhibited flexural strengths of 29% and 34.8%, respectively, significantly increasing over the unfilled cr-Ep. In addition, the flexural modulus of these nanocomposites has increased by 35.9% and 33.8%. Furthermore, compared to GnP/cr-Ep, HnT/cr-Ep and M-cnT/HnT/cr-Ep nanocomposite, M-cnT/cr-Ep and M-cnT/GnP/cr-Ep composite's Izod impact energy produced superior impact resistance. The distinct structures of M-cnT and GnP considerably improved the comprehensive performance of M-cnT and hybrid reinforced M-cnT/GnP into cr-Ep composites, and the insertion of these nanoparticles realizes the effective bonding between nanofillers and cr-Ep. This work offers a novel and practical method for improving the bonding between the nanofiller and the cr-Ep composite.

Keywords - Carboxyl-terminated butadiene-acrylonitrile-modified epoxy, Graphene nanoplatelets, Multiwalled carbon nanotubes, Halloysite nanotubes, Mechanical properties.

1. Introduction

A class of technical polymeric material systems known as engineering polymers can be used at temperatures ranging from 100°C to 150°C in various industrial settings. These polymers have good chemical resistance, robust mechanical and thermal stability, great dimensional stability, and wear resistance. Moreover, due to their low cost, repeatability, and ease of processing, these polymers quickly rise to the top of the materials sought for engineering applications. Over the past few decades, there has been a notable growth in research on polymer-based nanocomposites due to the development of sophisticated materials for potential uses.

Polymer nanocomposites, which contain carbon nanofillers, have a plethora of potential applications in thermal management, fuel cells, electronics, and transportation. Because of their exceptional properties and distinctive nanostructures, carbon base nanofillers, such as Carbon nanotubes (CnT) and Carbon nano fibers (CnF), can be added to a variety of polymers to enhance their mechanical, electrical, and thermal conductivities [1-6]. Shiu and Tsai [4] investigated the mechanical properties of the graphene/epoxy nanocomposites in terms of three distinct graphene, including Young's modulus, glass transition temperature, and coefficient of thermal expansion.

According to the mechanical and thermal properties, the nanocomposites, including intercalated graphene, had a greater glass transition temperature, a lower thermal expansion coefficient, and a higher Young's modulus than those containing graphene flakes. This is because intercalated graphene has the potential to increase the amount of high

density polymer in the nanocomposites and improve their overall characteristics. Using Graphene platelets (GnP), Single-walled Carbon nanotubes (s-CnT), and Multi-walled carbon nanotubes (M-cnT) at a nanofiller weight fraction of 0.1%, Rafiee et al. [5] examined the mechanical characteristics of Epoxy (Ep) nanocomposites. The findings show that GnP performs noticeably better than CnT. In contrast to a 3% increase for s-CnT, the GnP nanocomposite Young's modulus was 31% higher than that of the pure Ep. Using GnP increased the pristine Ep's tensile strength by 40%, while M-cnT only improved it by 14%.

The improved mechanical property increase of GnP over CnT could be attributed to their large specific surface area, enhanced adhesion/interlocking of nanofiller-matrix resulting from their wrinkled surface, and planar shape. By adding 0.1 wt% of Graphene Nanosheets (GNS) and GnP to the Ep, Shokrieh and colleagues [6] showed an increase in tensile strength of 15.7% for GNS and 14% for GnP. Higher weight percentages of nanoparticles in Ep also increased Young's modulus. Nevertheless, there are two primary obstacles to the application of CnF/polymer nanocomposites: firstly, their cost is high, and secondly, their poor dispersion within the polymeric matrix restricts the amount of possible advancement in polymer nanocomposites that may be achieved. These are important considerations when employing polymer-based composites integrating CnF [1].

Because of its exceptional mechanical properties, thermal stability, solvent resistance, and simplicity of processing, Ep, in particular, is a thermoset resin that is widely utilized in the aerospace, automotive, and marine industries [8, 9]. Epoxybased nanocomposites are another excellent matrix for creating nanocomposites with several potential uses, including fuel cells, electronics, thermal management, and transportation, especially when combined with carbon nanofillers [9, 10].

Carbon nanotubes (cnT) are a unique form of crystalline carbon that comes in two varieties: single-walled and multiwalled. As a result of their adaptable mechanical and physical characteristics, they have attracted a great deal of interest from researchers recently. CnT is an excellent choice for the next generation of composites, with tensile strengths between 50 and 100 GPa and an elastic modulus between 500 and 1000 GPa [11]. Comprehensive research has been conducted on the cnT reinforced composites using a range of matrix materials, such as metals [12], ceramics [13], and polymers [14].

Breton [15] evaluated the tensile properties of M-cnT /Ep composites by dispersing various kinds of catalytically produced M-cnT in Ep resin. The outcomes showed that Young's modulus of the Ep containing 1 wt%, 3 wt%, and 6 wt% M-cnT increased. Zhou [16] investigated the effects of

CnT loading on the tensile properties of Ep nanocomposites with 0.1 wt% to 0.4 wt%. The modulus rose as the CnT content increased, and the Ep composite loaded with 0.3 wt% CnT had the highest strength based on experimental findings. According to Allaoui's research [17], the composite with 1 wt% M-cnT had a yield strength and an elastic modulus that were twice as high as those of the pristine Ep, while the Ep composite with 4 wt% M-cnT showed the highest strength and modulus. Graphene is a possible reinforcement in nanocomposites due to its huge specific surface area and 2D layer structure, in addition to its exceptional mechanical properties. Graphene sheets and Ep resin have been mixed in a number of investigations to form nanocomposites, and the results indicate that the modulus and glass transition temperature of these materials can be greatly increased [18– 24]. Graphene is the strongest material ever measured, according to experiments by Lee and colleagues [22]. They also show that mechanical testing may allow atomically flawless nanoscale materials to withstand deformations beyond the linear regime. Shen et al. [23] investigate the reinforcing effect of graphene in improving the cryogenic tensile and impact performances of Ep composites at a weight fraction of 0.05–0.50%.

The findings demonstrate that while graphene aggregation occurs and gets more severe as graphene content increases, the graphene dispersion in the Ep matrix is good at low values. Furthermore, incorporating graphene at the appropriate concentrations significantly increases the composites' cryogenic tensile and impact strengths at liquid nitrogen temperature. Tang et al. investigated the impact of the graphene dispersion state on the mechanical characteristics of graphene/Ep composites [24].

Reduced Graphene Oxide (RGO) sheets were dispersed in various ways, with and without ball mill mixing. Although there were no appreciable variations in the tensile and flexural moduli due to the varying dispersion levels, they discovered that the composites with well scattered RGO had higher glass transition temperatures and strengths than those with weakly dispersed RGO. Also, Tang et al. [24] looked into the effects of graphene dispersion on the mechanical properties of graphene/Ep nanocomposites. It was demonstrated that highly distributed graphene nanocomposites have greater strength and fracture toughness. The mechanical performances of Ep resin reinforced with GnP, S-cnT, and M-cnT were compared by Rafiee et al. [5]. It was verified that the nanocomposites combined with GnP exhibit greater strength for the same quantity of reinforcement.

Researchers have studied various aspects of hybrid nanofiller reinforced polymer composites [25-31]. Hybrid nanofillers i.e., [montmorillonite/](https://www.sciencedirect.com/topics/earth-and-planetary-sciences/montmorillonite) cnT a review [25], M-cnT /synthetic diamond and M-cnT/boron nitride [26], clay/ McnT [27], M-cnT and GnP [28] silica/reduced graphene oxide [29], CnT/clays [30], CnT/graphene [31] in various polymer nanocomposites characterization resulted in considerable enhancement of mechanical properties.

The mechanical properties of hybrid nanofiller-filled composites have also been the subject of numerous investigations. Adding nanoparticles can easily enhance Young's modulus of polymer matrices [32]. The stress transfer between the nanoparticles and the matrix significantly impacts strength. The applied stress can be efficiently transferred to the particles from the matrix for well-bonded nanoparticles [33], improving strength. On the other hand, adding particles reduces the strength of weakly bound nanoparticles.

A rapidly developing field of study can potentially improve the mechanical, thermal, electrical, and barrier qualities of materials based on epoxy by incorporating nanofillers into epoxy resins. Despite significant breakthroughs, there are still a number of research gaps in this field. Some major gaps are processing methods, homogeneous nanofiller dispersion, interfacial bonding, and multifunctional characteristics. It will take a multidisciplinary strategy to close these research gaps, integrating various nanofillers with appropriate processing methods. The novelty of the current research work is that more research and development in these areas will open the door for the next generation of highperformance epoxy composites. It is also evident from the literature that modest nanofiller loadings are necessary to improve the performance of polymer-based nanocomposites. Furthermore, it was found that certain mechanical characteristics increase with $<$ 3 wt% of nanofillers. The processing parameters that are employed have a considerable impact on the properties and interfacial characteristics of polymer-based nanocomposites.

Consequently, great consideration must be given to selecting the appropriate processing technique, parameters, and chemical treatment of nanofillers to achieve the desired nanocomposite qualities. Nevertheless, an examination of the already available literature indicates that the interactions among the hybrid nanofillers have not received much attention, especially when it comes to mechanical properties. The problem of hybrid silane treated nanofillers when carboxyl-terminated butadiene-acrylonitrile-modified Epoxy (cr-Ep) combined with M-cnT/GnP and M-cnT/HnT is thus addressed in the current study. Various mechanical characteristics were examined, including tensile strength, Young's modulus, flexural strength, flexural modulus, and Izod impact strength.

2. Materials and Methods

2.1. Materials

Epoxy (Ep) resin with medium viscosity (Lapox L-12) and light-yellow aliphatic polyamine (Lapox K-6) were the thermosetting matrix materials used in this research work. They were bought from Atul India Private Limited, Gujarat,

India. The density of the epoxy resin is 1.17 g cm⁻³. The source of dimethylformamide was from Spectrochem, India. Triphenylphosphine (molecular weight: 262.29 g mol⁻¹) and Carboxyl-Terminated acrylonitrile Butadiene copolymer (CTBN; molecular weight: 3600 g mol-1) were purchased from Sigma Aldrich, Bangalore, India. Merck India Pvt Ltd, Bangalore, is the source of the reinforcing agents, which include Multiwalled carbon nanotubes (M-cnT), Graphene nanoplatelets (GnP), and Halloysite nanotubes (HnTs). The GnP have a material density of 0.2 to 0.4 g cm⁻³ and three to six layers, each with a surface area of $750 \text{ m}^2 \text{ g}^{-1}$. The M-cnT had a diameter of 30 nm, an average length of 10 mm, and a density of 2.1 $g \text{ cm}^{-3}$. They were produced using catalytic chemical vapor deposition. The HnTs are spherical, with an average particle diameter of 30 to 70 nm and lengths ranging from 1 to 3 μ m. HnTs have a surface area of 64 m² g⁻¹ and a density of 2.53 g cm⁻³.

2.2. Fabrication

2.2.1. Preparation of CTBN Modified Epoxy

The necessary amount (10 wt%) of liquid CTBN rubber (cr) was combined with Epoxy (Ep) resin and stirred mechanically for 50 min at 80°C. Subsequently, the mixture was degassed in a vacuum oven and ultrasonicated to release any retained air bubbles.

Once the liquid had cooled to room temperature, the stoichiometric amount of the curing agent was added and manually stirred for 10 min. After that, the mixture was put into moulds and, as per the manufacturer's recommendation for neat Ep, cured for 24 h at 25° C and post-cured for 2 h at 100 $^{\circ}$ C. The Ep that had been altered by cr was coded as cr-Ep.

2.2.2. Preparation of CTBN Modified Epoxy Nanocomposites

The creation and mixing of five different kinds of CTBNmodified Epoxy (cr-Ep) based nanocomposites is depicted in Figure 1. The process used to create the cr-Ep based nanocomposites with a loading of 1 wt% of M $cnT/GnP/HnT$ and hybrid filler loadings; 0.5 wt% M-cnT + 0.5 wt% GnP and 0.5 wt% M-cnT + 0.5 wt% HnT. After 20 min of sonication, the necessary amount of nanofiller was mixed in an acetone combination. This mixture was then mixed with the cr-Ep matrix. The fluid underwent another 20 min of sonication to achieve a homogeneous dispersion of nanoparticles. It took 50 min to remove the acetone by heating it to 50 °C. The tiny amount of solvent was removed using a vacuum oven. It was added to the K-6 hardener (12 g/100 g of cr-Ep), stirred for 10 min, and then given 15 min to degas. The cr-Ep and its mono and hybrid nanocomposites were made with a rectangular mold measuring $150 \text{ mm} \times 200 \text{ mm} \times 3$ mm. The mixture was allowed to cure for 24 hours at room temperature after being placed into the mold. Two hours of post-curing were spent at 100° C. The same method was used to prepare the hybrid nanocomposites of cr-Ep. The synthesized mono and hybrid nanocomposites, their codes, and complete compositions are listed in Table 1.

Fig. 1 Schematic of the dispersion of nanofiller in cr-Ep via solution mixing, ultrasonic agitation, high-speed shear mixing and preparation of nanocomposites

2.3 Mechanical Testing

2.3.1. Tensile Test

Tensile tests were performed using an automated Universal testing machine (Capacity: 100 kN). Kalpak Instruments and Controls Pune, India, made this machine. For this testing, a constant deformation rate of 25 mm/min and a constant room temperature of 23°C were employed. As per ASTM D638-17, all data were recorded for the 55 mm tested gauge length.

In compliance with ASTM standards [34], dog boneshaped specimens were used to record the results of tensile tests. Five tests were conducted, and the measured data's mean values and standard deviations were computed.

2.3.2. Flexural Test

Using a computerized universal testing machine made by Kalpak Instruments and Controls, Pune, India, with a maximum load cell capacity of 100 kN. Three-point bending tests were performed as per ASTM D790- 17 [35].

The mean and standard deviations of the measured data were considered, and each experiment was carried out five times. The coupons were maintained at a constant span to depth ratio of 20:1, and a 10 kN load cell with 2.5 mm/min cross head speed was selected for flexural testing.

2.3.3. Izod Impact Test

Following ASTM D256-10 [36], the Izod impact tests were carried out using an R2 hammer on an impact testing machine (Make: International Equipments, Mumbai, India) with an impact energy capacity of 0-5.24 J. Using a notch cutter, a 45° notch was made in the specimen, limiting its width to 10.16 mm. For every series of composites, 5 specimens measuring 64 mm **×** 12.5 mm were put to the test.

3. Results and Discussion

The goal of adding nanofillers is to improve the epoxy mechanical properties of CTBN-modified epoxy (cr-Ep). The objectives of the nanofillers like M-cnT, GnP, and HnT have been met, as depicted in Figures 2, 3, 4, 5, and 6.

3.1. Role of Nanofillers on Tensile Properties of Epoxy

To study the synergetic effect of the tensile properties for cr-Ep nanocomposites, we compared M-cnT, GnP, and HnT at a fixed 1 wt%. Experimental data from Table 2 and the graphical illustration in Figure 2 make it clear that the Tensile Strength (TS) is higher than unfilled cr-Ep regardless of the kind of nanofillers used. Tensile strength was enhanced by 32.2% when M-cnT was incorporated into cr-Ep compared to unfilled cr-Ep. Similarly, adding GnP and HnT nanofillers to the cr-Ep matrix material improved the TS. The hybrid nanocomposite M-cnT/GnP/cr-Ep exhibited the highest TS value. The increase in TS for M-cnT/GnP/cr-Ep and McnT/HnT/cr-Ep hybrid nanocomposites are 39.5% and 27.6% respectively. Moreover, the hybrid nanofiller changed the cr-Ep and demonstrated a supportive impact in raising the TS.

It is evident from Figure 2 that the stress and strain values for all mono and hybrid nanocomposites are significantly influenced by the kind and loading of nanofillers. The occasionally observed low reinforcing ability of the nanofillers in epoxy nanocomposites can be attributed to a variety of factors, including poor dispersion and a lack of interfacial adhesion, both of which are essential for load transfer in composites. According to Bokobza [37], the smoothness of the cnT surfaces may restrict the load transfer from the matrix to the cnT reinforcement. According to Gojny et al. [38], insufficient dispersion of cnTs in the polymer matrix caused load transfer to be restricted, which is important for creating reinforced nanocomposites. Smutisikha [39] determined that cnTs have a strong tendency to agglomerate due to their high aspect ratio and large surface area, which causes inhomogeneous dispersion in the polymer matrix.

Table 2. Tensile test results of cr-Epoxy nanocomposites

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Nanocomposites	Strength	Modulus	Strain			
	(MPa)	(MPa)	$($ %)			
$cr-Ep$	69.3 ± 3.8	$1889+8.8$	3.19 ± 0.2			
M -cn T	91.6 ± 3.1	$3899+9.8$	$2.99+0.1$			
GnP	86.3 ± 3.4	$3250+5.8$	2.82 ± 0.3			
HnT	83.5 ± 3.7	2980±7.8	2.51 ± 0.2			
M -cnT/GnP	96.7 ± 3.3	4124 ± 6.8	2.91 ± 0.1			
$M-nT/HnT$	$88.4 + 3.5$	3485 ± 4.8	2.75 ± 0.2			

Fig. 2 Tensile strength and tensile modulus of cr-Ep and their mono and hybrid nanocomposites

Fig. 3 Tensile strain of cr-Ep and their mono and hybrid nanocomposites

The tensile properties cannot be determined without considering the Tensile Modulus (TM). The tensile test results unequivocally show that, compared to unfilled cr-Ep, the TM of M-cnT/cr-Ep rose by 105.9%. The addition of GnP and HnT fillers resulted in a rise in TM. The TM of GnP/cr-Ep and HnT/cr-Ep had grown by 72.1% and 57.8%, respectively, compared to unfiled cr-Ep.

An increase of 57.8% was observed when comparing McnT and GnP filled mono nanocomposites to HnT/cr-Ep; however, this was not as significant as what was observed with hybrid filler filled cr-Ep composites. Both TS and TM improved considerably with hybrid nanofiller loading of McnT and GnP reinforced cr-Ep nanocomposites. With a TM of 118,3%, the hybrid nanocomposite M-cnT/GnP/cr-Ep showed the highest TM.

However, we observed a little enhancement in the tensile characteristics with HnT loading (Figure 2). All of the nanofillers examined in this work enhance the TM of cr-Ep by decreasing the flexibility of the polymer chains. Free volume space decreases due to attractive polar interactions and Van der Waals bonds forming between cnTs and epoxy chains. The epoxy chains carry extra loading due to the M-cnTs effect, as per Park et al. [40].

The improvement in the tensile characteristics of the cr-Ep based mono and hybrid nanocomposites in this work is due to the strong interfacial adhesion between nanofillers and cr modified Ep and the uniform filler dispersion. The earlier research findings align with the literature [41-43].

According to the findings, M-cnT and GnP were more important in the cr-Ep nanocomposites. In the cr-Ep matrix, M-cnT and GnP hybridization (M-cnT/GnP), GnP, in particular, played a more effective reinforcement role, as shown in Figure 2 and Table 2. The incredibly large specific surface area of the GnP, which carried significant amounts of stress transfer across the interface and offered more effective reinforcement than that of M-cnT, could be one reason for this behavior.

Figure 3 shows the strain at fracture for cr-Ep and their mono and hybrid nanocomposites. Figure 3 makes it clear that for cr-Ep, 3.19% strain was seen before the matrix began to elongate without a discernible rise in the axial load. At this

stage, there is a decrease in the lateral dimension and an increase in the longitudinal dimension of the predominant elastic nature. Furthermore, the strain dropped to 2.99%, 2.82%, and 2.51%, respectively, with the addition of 1 wt% M-cnT, GnP, and HnT fillers. Furthermore, a reduction in strain was found in M-cnT/GnP and M-cnT/HnT filled cr-Ep hybrid nanocomposites at 2.91% and 2.75%, respectively. In this work, the decreased elasticity caused by the nanofillers leads to the brittleness of mono and hybrid composites. Deshmukh and Joshi have attributed the decrease in strain at fracture and the enhancement in tensile properties to the interlaminar strength and stress transfer [44]. Figure 3 shows that when the nanofillers were incorporated into cr-Ep, the material's brittleness predominated, and its elastic nature was diminished, causing the material to fail at a lower strain value as its load carrying capacity increased.

3.2. Effect of Nanofillers on Flexural Properties of Epoxy

According to ASTM D790, the post-cured cr-Ep and their mono and hybrid nanocomposites were measured for Flexural Strength (FS) and Flexural Modulus (FM). The variation in

flexural strength and flexural modulus is displayed in Figure 4 and summarized in Table 3. Tensile, compression and shear strengths combine to provide the composites' flexural properties. The flexural characteristics of composites are typically enhanced by high elastic fillers [45]. It is evident from Figure 4 that improving the FS and FM of the cr-Ep composites results from using all nanofillers. For the hybrid nanofillers in the cr-Ep composites (M-cnT/GnP and McnT/HnT), both FS and FM are markedly enhanced.

Nano composites	Strength (MPa)	Modulus (MPa)	л (%)	Impact strength
				\mathbf{J}
$cr-Ep$	$96.5 + 4.5$	2845 ± 8.5	7.2 ± 0.2	0.18 ± 0.2
M -cn T	$124.5 + 3.8$	3865 ± 7.4	$7.1 + 0.1$	0.25 ± 0.1
GnP	$121.9 + 3.1$	$3724 + 7.5$	6.9 ± 0.2	$0.22+0.1$
HnT	112.3 ± 4.2	$3289 + 7.1$	6.6 ± 0.3	0.19 ± 0.1
M -cnT/GnP	130.1 ± 4.2	3805 ± 6.5	$7+0.1$	$0.24 + 0.2$
$M - cnT/HnT$	$119.8 + 4.4$	$3678 + 8.5$	6.8 ± 0.2	$0.21 + 0.3$

Table 3. Flexure and impact test results of cr-Epoxy nanocomposites

Fig. 4 Flexural strength and flexural modulus of cr-Ep and their mono and hybrid nanocomposites

Fig. 5 Deflection at break of cr-Ep and their mono and hybrid nanocomposites

The results show that the FS of the M-cnT/GnP modified hybrid nanocomposite increased by approximately 34.8% and 33.8%, whereas the FS and FM of the M-cnT/HnT modified hybrid cr-Ep nanocomposites increased by 24.2% and 29.3%. Compared to all other composites examined, the impact of hybrid nanofillers, specifically M-cnT/GnP, is more noticeable in improving the FS and FM of cr-Ep. It is possible that better interfacial interaction between c-Ep and nanofillers, as well as uniform dispersion and good adherence, are the reasons for the notable increase in FM and FS of mono and hybrid nanocomposites. The stiffness of high elastic modulus fillers may impact the FM of polymer matrix composites [46]. Because M-cnT could entangle Ep chains more effectively than GnP and HnT nanoparticles, producing a greater restriction to the alignment of Ep chains, they could give more enhancement in FM. The outcomes correlate with the research conducted by Kozlov et al. [46] and Chaturvedi et al. [45]. Referring to Figure 4, one can quickly identify that the superior FS and FM of the mono and hybrid filler-modified cr-Ep nanocomposites may be attributed to a combination of factors, including high modulus, effective load transfer, good dispersion, and strong interfacial interactions.

Variations in the failure deformation (Δ) of nanocomposites based on cr-Ep are shown in Figure 5 in relation to different nanofillers. The figure clearly shows that the deflection at the failure of mono and hybrid nanocomposites is smaller than that of unfilled cr-Ep, containing a 7.2% deflection at the break. Figure 5 clearly shows that the matrix containing cr-Ep began to distort before a discernible rise in the bending force. Furthermore, the deflection at break decreased by 7.1%, 6.9%, and 6.6%, respectively, when 1 wt% M-cnT, GnP, and HnT fillers were included in cr-Ep. Moreover, deflection at break was observed in M-cnT/GnP and M-cnT/HnT filled cr-Ep hybrid nanocomposites at 7% and 6.8%, respectively. Consequently, cr-Ep based nanocomposites with 1D and 2D fillers could somewhat minimize deflection at failure compared to unfilled cr-Ep. This illustrates even more how, as seen in Figure 5, every nanofiller used in this study could more effectively restrict the movement of the chains in the Ep matrix.

Generally speaking, nominal properties like the anisotropy of the nanoparticles and polymer matrix composite's elastic modulus are used by micromechanical models to characterize the elastic modulus of polymer based nanocomposites. Particle aggregation causes the true elastic modulus of the nanofiller to deviate significantly from its stated value. Kozlov et al. [46] investigated the modulus of the nanoparticles aggregate in polymer based composites strengthened with particles (0-D), CnT (1-D) and graphene (2- D) nanofillers. They found that the fractal dimension was the most physically accurate and broadly applicable parameter of the particle aggregates in various nanofillers. The polymer matrix's stiffness and the particle aggregates' structure determined their true elastic modulus. Coleman et al.'s research [47] showed that, despite CnT's nominal elastic

modulus of around 1000 GPa, its actual elastic modulus in polymer matrix composites was 71 ± 55 GPa. Jelic et al. [48] investigated the possibility of HnT to improve the Ep matrix's mechanical characteristics and thermal stability. The obtained results demonstrated that the distinctive properties of welldispersed HnT, agglomeration scale, and reduced void presence could be controlled during the production process to produce HnT/Ep nanocomposites with enhanced characteristics. Darshan et al. [49] developed biocompatible, lightweight polymer-based nanocomposites for load-bearing applications. They investigated the effects of HnT loading on the wear behavior and mechanical characteristics of Ep composites reinforced with basalt and silk fibers. In this work, HnT is added to basalt and silk fibers to reinforce Ep composites, improving their mechanical and tribological properties.

3.3. Role of Nanofillers on Impact Energy of Epoxy

To ascertain the composite sample's capacity to absorb impact energy during the crack's propagation, tests were conducted on unfilled cr-Ep and their nanocomposites. The impact energy of pristine cr-Ep and its mono and hybrid fillerfilled composites are displayed in Figure 6, and mean data are listed in Table 3. The addition of each individual M-cnT, GnP, and HnT particles to cr-Ep has increased the impact strength.

It is commonly recognized that adding nanofillers will increase the polymer matrix's resistance to impact load. Finally, we saw that all of the nanocomposites had greater impact strength. For M-cnT, GnP, and HnT reinforced cr-Ep mono composites, the impact strength increases in the following order: 0.25 J, 0.22 J, and 0.19 J. Additionally, the hybrid loading of M-cnT/GnP and M-cnT/HnT nanofillers into cr-Ep results in 0.24 J and 0.21 J increase in impact strength. This increase is explained by the nanofillers' better dispersion inside the cr-Ep matrix material. This limits the direction of fracture flow transverse to the nanofillers. Additionally, nanoparticles' role as stress concentration sites during loading encourages cavitations at the particle-matrix boundaries. The enhanced impact strength in the GnP/cr-Ep mono composite is explained by the robust interfacial interaction between GnP and the cr-Ep matrix's epoxide group. These cavitations may release the plastic constraints and cause the matrix to deform plastically in bulk, which could lead to improved impact energy. An increase in impact strength was observed when M-cnT wt% increased due to the reinforcing effect of M-cnT on the Ep matrix phase [50]. This effectively illustrated how the toughness and high impact properties of M-cnT increased the impact strength of the nanocomposite by facilitating the efficient passage of stress from the cr-Ep matrix to the M-cnT and improving the impact strength of M-cnT/Ep. Furthermore, the impact strength increased with the addition of M-cnT, GnP, and HnT nanofillers, as seen in Figure 6, because of enhanced toughness and epoxy matrix bonding. Comparable outcomes have been reported for hybrid nanocomposites [7, 50–52].

Fig. 6 Izod impact strength of cr-Ep and their mono and hybrid filler filled nanocomposites

4. Conclusion

In this research article, the investigation was performed on the mechanical behaviour of M-cnT, GnP, HnT, and hybrid fillers consisting of M-cnT/GnP and M-cnT/HnT reinforced CTBN modified epoxy nanocomposites and portrayed an improvement in mechanical properties upon reinforcement.

- The tensile strength and modulus of the M-cnT/GnP/cr-Ep hybrid nanocomposite were much higher than those of the unfilled cr-Ep, at 39.5% and 118.3%, respectively. Therefore, a change in contact geometry from 0-D point contact to 1-D/2-D linear contact, which significantly increases the contact surface area inside the cr-Ep matrix material, is responsible for the synergistic effect.
- The M-cnT/GnP reinforced cr-Ep had the best flexural strength and the highest flexural modulus compared to unfilled cr-Ep and other mono and hybrid nanocomposites, significantly improving flexural characteristics. The increased mechanism was elaborated in the epoxy nanocomposites treated with CTBN due to the larger interaction energy between the nanofiller and surrounding matrix.
- Compared to unfilled cr-Ep and HnT/cr-Ep nanocomposite, the impact resistance of nanofillers such

as M-cnT and GnP reinforced cr-Ep nanocomposites is higher. Because M-cnT and GnP attenuate more energy than HnT, the impact strength findings indicate that GnP filler and M-cnT also substantially absorb impact resistance.

Ultimately, this study has demonstrated that the mechanical characteristics of CTBN rubber modified epoxy may be improved by the synergistic effects of M-cnT and GnP hybrid nanofillers at low content (0.5 wt% M-cnT and 0.5 wt% GnP). More research must be done to understand the mechanical characteristics of hybrid nanocomposites at various loadings and settings. Furthermore, the work's findings suggest that M-cnT and GnP nanofillers are promising for structural reinforcing additives in epoxy hybrid nanocomposites modified with CTBN rubber.

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