

Mechanical Properties of SLJs with Graphene and MWCNT Nanoparticle-Doped Hybrid Polyurethane Adhesives on Epoxy-Based Carbon Fiber Reinforced Composite Plates

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ABSTRACT

Keywords:

Mechanical Properties,
Tensile Test,
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Polyurethane Adhesives,
Scanning Electron Microscopy

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In this study, we investigated the mechanical effect of by weight of 0.05%, 0.1% and 0.15% Graphene Nanoplates (GNPs) and MWCNTs doped polyurethane adhesives on single lap joint of Epoxy based and Carbon Fiber Reinforced Composite (CFRP) plates. First of all, Nanocomposite adhesive was produced by adding different weight ratios of nanoparticles into polyurethane adhesive. Then, the tensile test samples were produced using these adhesives and tensile tests were performed in accordance with the ASTM D 882 standard. According to the obtained results, the additive ratios showing the best mechanical properties were determined and single lap joints (SLJ) were produced. Then, the mechanical properties of the SLJ were tested in accordance with the ASTM D1002-10 standard under a constant tensile loading ratio. The highest tensile strength was observed in the 05GR15CNT sample with an increase of 52.67% (14.58 MPa) among the tensile test specimens. On the other hand, the highest shear strength was obtained in the 10GR10CNTSLJ sample with an increase of 14.80% (15.51 MPa) for the SLJ specimens. The distribution of the nanoparticles and the morphology of the failure surfaces were analyzed by scanning electron microscopy (SEM) images.

Grafen ve MWCNT Nanoparçacık Katkılı Poliüretan Yapıştırıcı ile Birleştirilmiş Epoksi Bazlı Karbon Fiber Takviyeli Kompozit Levhaların Tek Taraflı Bindirmeli Bağlantılarının Mekanik Özellikleri

ÖZ

Bu çalışmada, epoksi esaslı karbon fiber takviyeli kompozit (CFRP) plakaların tek taraflı bindirme bağlantılarda, ağırlıkça %0.05, %0.1 ve %0.15 grafen nanoparçacık (Gr) ve çok duvarlı karbon nanotüp (ÇCKNT) katkıli poliüretan yapıştırıcıların mekanik etkisi incelenmiştir. İlk olarak, farklı ağırlık oranlarında nanoparçacıklar poliüretan yapıştırıcıya eklenerek nanokompozit yapıştırıcı üretilmiştir. Daha sonra, bu yapıştırıcılar kullanılarak çekme testi numuneleri üretilmiş ve çekme testleri ASTM D 882 standardına uygun olarak gerçekleştirilmiştir. Elde edilen sonuçlara göre en iyi mekanik özellikleri gösteren katkı oranları belirlenmiş ve tek taraflı bindirme bağlantıları (TTBB) üretilmiştir. Ardından, TTBB'nin mekanik özellikleri sabit çekme yükleme oranı altında ASTM D1002-10 standardına uygun olarak test edilmiştir. Çekme test numuneleri arasında en yüksek çekme dayanımı, %52.67 artışla (14.58 MPa) 05GR15CNT numunesinde gözlemlenmiştir. Öte yandan, TTBB numuneleri arasında en yüksek kayma dayanımı, %14.80 artışla (15.51 MPa) 10GR10CNTSLJ numunesinde elde edilmiştir. Nanoparçacıkların dağılımı ve hasar yüzeylerinin morfolojisi, taramalı elektron mikroskobu (SEM) görüntüleri ile analiz edilmiştir.

Anahtar Kelimeler :

Mekanik Özellikler,
Çekme Testi,
Nano Parçacık Katkılı
Poliüretan Yapıştırıcılar,
Taramalı Elektron Mikroskobu

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1. Introduction

Adhesive is defined as a chemical bonding material applied to the surfaces of materials to bind and hold them together [1]. Adhesive bonds are preferred due to their advantages such as addressing the shortcomings of traditional joining methods, combining different materials, and providing uniform stress distribution [2]. A range of adhesive types, including polyurethane, epoxy, acrylic, silicone, and latex, are available for diverse structural applications [3]. Polyurethane adhesives (PU) are essential in numerous industries because of their versatility and superior performance attributes [4]. PU adhesives are utilized in various applications, thanks to their outstanding resistance to external factors, including as coatings, binders for efficient processes like seawater desalination, sealing components, foam in sandwich panels, adhesives for industrial purposes, and even in biomedical applications [5]. Due to their extensive range of applications, various methods have been proposed to increase the mechanical properties of structural adhesives. It has been observed that the strength of adhesive joints depends on the type and surface quality of the adherend, the application and curing techniques of the adhesive, the size of the adhesive joints, testing under different loading conditions, and environmental factors such as temperature and humidity [6]. In addition to these properties, the incorporation of nanoparticle reinforcements into adhesives has emerged as a prominent method for enhancing the strength of adhesive bonds [7]. Nanoparticles, such as aluminum oxide (Al₂O₃), silica (SiO₂), nano clay, rubber particles, carbon black, graphite, GNPs, MWCNTs, and fullerenes, are advanced materials that offer a wide range of opportunities for obtaining nanocomposite materials [8]. MWCNTs and GNPs with exceptional properties have gained considerable recognition among researchers. Numerous studies have investigated the effects of these components in detail [9]. MWCNTs have an extremely high aspect ratio and are regarded as one-dimensional nanomaterials with exceptional mechanical properties. Their unique structure and outstanding mechanical and physical properties render them advanced fillers for the development of new composite adhesives [10]. Gilad Otorogust et al. [11] developed a nanocomposite adhesive by adding MWCNTs at concentrations of 0.1 to 0.4 wt% to structural polyurethane (PU) adhesives with the aim of improving their properties. According to SLJ shear test results, the bonded shear strengths increased by up to 64% and 22%, respectively. Wernik et al [7] conducted tensile tests on dogbone specimens, tensile bond tests, double lap shear tests, and double cantilever beam fracture toughness tests to experimentally investigate the mechanical properties of MWCNT-reinforced epoxy adhesives. Experimental observations indicate that the greatest improvement in measured properties occurs at a critical carbon nanotube concentration of approximately 1.5 wt%. However, at concentrations exceeding this critical value, the properties begin to degrade, sometimes falling below the levels of pure epoxy. Ozkan et al [12] investigated the effects of nanoparticle hybridization on the shear and flexural performance of SLJs of glass fiber reinforced polymer (GFRP) composites. For this purpose, MWCNT and silica nanoparticles were incorporated into the epoxy adhesive at various concentration rates. The effects of these nanoparticles on adhesion performance under different loads were analyzed through three-point bending tests and single-lap shear tests. The maximum shear and flexural strengths were achieved with the combination of 0.5 wt.% MWCNT and 0.25 wt.% nano-silica particles, showing improvements of 45.4% and 63.2% respectively, compared to pure samples. Scanning electron microscopy (SEM) was used to examine fracture mechanisms and failure modes, revealing that nanoparticle-doped samples exhibited higher load-bearing capability, with observed failure mechanisms including crack deviation, crack pinning, pull-out and bridging. Jia et al [13] conducted an experimental study to investigate the mode I fracture resistance of epoxy construction adhesive reinforced with graphene nanoplatelets (GNPs) using double cantilever beam (DCB) samples. The study revealed that the incorporation of 0.25 wt% graphene resulted in a fivefold enhancement in mode I fracture toughness compared to the neat epoxy adhesive. However, increasing the graphene content further led to a decrease in fracture toughness due to the aggregation of graphene particles within the adhesive matrix. Research studies often use low-viscosity laminating resins to facilitate the effective mixing and uniform dispersion of nanoparticles in adhesive formulations [14]. When nanoparticle concentrations are ≤ 0.5 wt%, significant variations in adhesive performance [15], particularly in terms of mechanical strength enhancement, have been reported [16].

In complex structures, it is often necessary to join components in a way that maintains structural integrity under different loads and environmental conditions. While metals are typically joined using techniques such as riveting, bolting, gluing, brazing, and soldering, the joining methods for polymer matrix fiber-reinforced composites are predominantly limited to adhesive bonding. This highlights the essential role of adhesives in the effective assembly of these advanced composite materials [17]. Research on the mechanical strength of adhesive joints focuses on factors such as joint geometry, overlap length, adhesive thickness, material properties, and environmental conditions. In recent years, studies on the effects of nanoparticles added to polyurethane adhesives on mechanical and thermal properties have been limited. The literature shows that nanoparticles are generally used in epoxy-based adhesives, while research on polyurethane adhesives remains scarce.

Nanoparticles enhance the thermal, electrical, and mechanical properties of adhesives while also improving environmental resistance and aging performance. Therefore, it is possible to develop hybrid adhesives by incorporating nanoparticles into polyurethane adhesives. Polyurethane was preferred in this study due to its cost-effectiveness and wide range of applications, and the potential of polyurethane-based hybrid adhesives has been demonstrated.

In this study, neat and nanoparticle-added polyurethane adhesives were used to join the epoxy-based carbon fiber-reinforced composite plates. Tensile tests for dog-bone adhesive samples and SLJ tensile tests for adhesively jointed composite plates were conducted to obtain the mechanical properties. SEM analysis was performed to examine the fractured surfaces after tensile tests.

2. Materials And Method

2.1. Materials

In this study, the adhesive used is KLB 75, a two-component polyurethane-based adhesive supplied by Duratek® company. This adhesive consists of polyester-based polyols used as catalysts in suitable proportions, accounting for 80% of the components, and a curing isocyanate mixture comprising the remaining 20%. The viscosity of KLB 75 adhesive at room temperature is 1800 mPas, with a density of 1.35 ± 0.10 g/cm³. The curing time at room temperature when used in the proportions specified by the manufacturer (80% polyol by weight, 20% isocyanate) is approximately 1 to 2 hours. Graphene and MWCNT were used as nanoparticles. The MWCNTs utilized have an outer diameter ranging from 15 to 25 nm, an inner diameter between 5 and 10 nm, and lengths varying from 10 to 20 μ m. The properties of graphene include a purity of 99.5%, a diameter of 24 μ m, a specific surface area of 150 m²/g, and an elasticity modulus of 0.5 TPa. The nanoparticles were procured from Nanografi company. In this study, CFRP plates produced by Kompozitsan company were used as the adhered material. The CFRP plates consist of 8 layers and have a total thickness of 2 mm.

2.2. Preparation of nanoparticles doped polyurethane adhesive

For the purpose of adhering the composite plates, nanoparticles doped adhesives were prepared. Different ratios of nanoparticles added into the polyurethane resin were mixed using a Bandelin HD 2200 ultrasonic mixer to ensure a homogeneous distribution without compromising the structure of the polyurethane and nanoparticles to prevent overheating and maintain the stability of the polyurethane-nanoparticle mixture, the mixing was performed in a beaker containing ice water. Then the solution was subjected to degassing at room temperature under a vacuum of 0.25 bar for one hour to evacuate any entrapped air bubbles within the solution. Subsequently, a hardening agent was added at a concentration of 20% and mechanically mixed for 10 minutes to ensure uniform dispersion. Following this procedure, the nano-adhesive was prepared for application.

2.3. Preparing the surfaces of adhesive bonding samples

In the surface preparation of CFRP plates, the bonding surfaces were abraded perpendicular to the direction of tension using 240-grid SiC sandpaper. Following abrasion, the samples were sequentially rinsed with tap water and distilled water, and subsequently immersed in acetone for 10 minutes. The samples were then dried in a sterilized oven at 60°C for 20 minutes to complete the surface preparation process.

2.4. Manufacturing of dogbone and single lap joints samples

The nanoparticle-doped and pure polyurethane adhesives, produced with nanoparticle additives, were poured into metal molds prepared according to ASTM D638 [18] standards as Figure 1, following the casting process, the dogbone tensile samples were left to cure completely at room temperature for 72 hours, in accordance with the manufacturer's recommendation. At the end of this waiting period, the samples were prepared for tensile testing. The nanoparticle type, additive percentage by weight (%), and abbreviated names of the prepared samples are as shown in Table 1. Using the manufactured nano-composite adhesives, CFRP-CFRP single lap joints were produced in accordance with the dimensions specified in ASTM D1002 [19] standards (Figure 1). During the preparation of bonded joints with the adhesive, a specially designed fixture was utilized to ensure the materials remained stationary and to adjust the desired adhesive thickness. After completing this process and placing the topmost sample, the fixture was closed, and the SLJs samples was subjected to a 72-hour curing process at room temperature.

Table 1. Naming of the produced nanocomposite materials

Samples	Additive by Weight (%)	
	Graphene	MWCNT
PU	-	-
05GR	0.05	-
10GR	0.1	-
15GR	0.15	-
05CNT	-	0.05
10CNT	-	0.1
15CNT	-	0.15
05GR05CNT	0.05	0.05
05GR10CNT	0.05	0.1
05GR15CNT	0.05	0.15
10GR05CNT	0.1	0.05
10GR10CNT	0.1	0.1
10GR15CNT	0.1	0.15
15GR05CNT	0.15	0.05
15GR10CNT	0.15	0.1
15GR15CNT	0.15	0.15

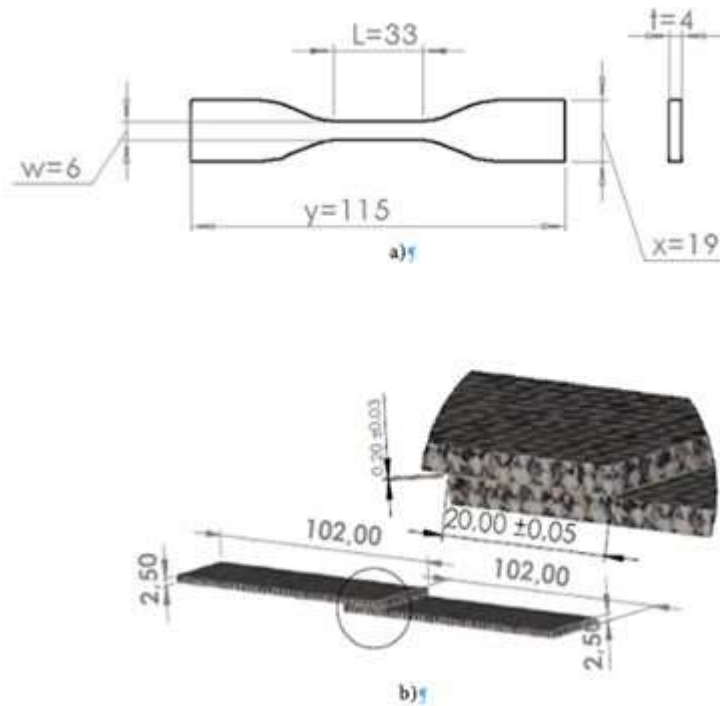


Figure 1. Schematic view of a) tensile sample, b) SLJ test sample

2.5. Characterization

Tensile tests of the dogbone samples were conducted according to ASTM D638, with the specimens loaded at a constant tensile speed of 2 mm/min until failure, while shear strength tests of composite-to-composite SLJs followed ASTM D1002 standards, with the SLJs tested at a tensile speed of 1 mm/min. Both tests were performed using a Shimadzu AGS-X tensile testing machine equipped with Trapezium-x software. An Epsilon 3560 model extensometer was employed to measure deformations in the adhesive regions of the single lap joint samples.

3. Experimental Results

3.1. Dogbone tensile tests results for nanocomposites

Tensile tests were conducted to determine the mechanical behavior of dogbone samples made from both doped and neat polyurethane adhesives, resulting in stress-strain curve (Figure 2). From these curves, the tensile strengths, maximum strains and toughness values of the nanoparticle-reinforced samples were determined. The results for tensile samples containing graphene nanoparticles at weight fractions of 0.05%, 0.10%, and 0.15% are presented in Table 2. The results for tensile samples containing MWCNT nanoparticles at weight fractions of 0.05%, 0.10%, and 0.15% are shown in Table 3. Additionally, the test results for hybrid adhesive tensile samples containing both graphene and MWCNT nanoparticles at weight fractions of 0.05%, 0.10%, and 0.15% are provided in Table 4. After the dog-bone tensile test, the tensile strength of the neat polyurethane sample was found to be 9.55 MPa. Compared to this, the maximum strain value was observed in the 05GR10CNT sample, with a 52.67% increase, reaching 14.58 MPa. The minimum strain value was recorded for the 10CNT sample at 7.57 MPa.

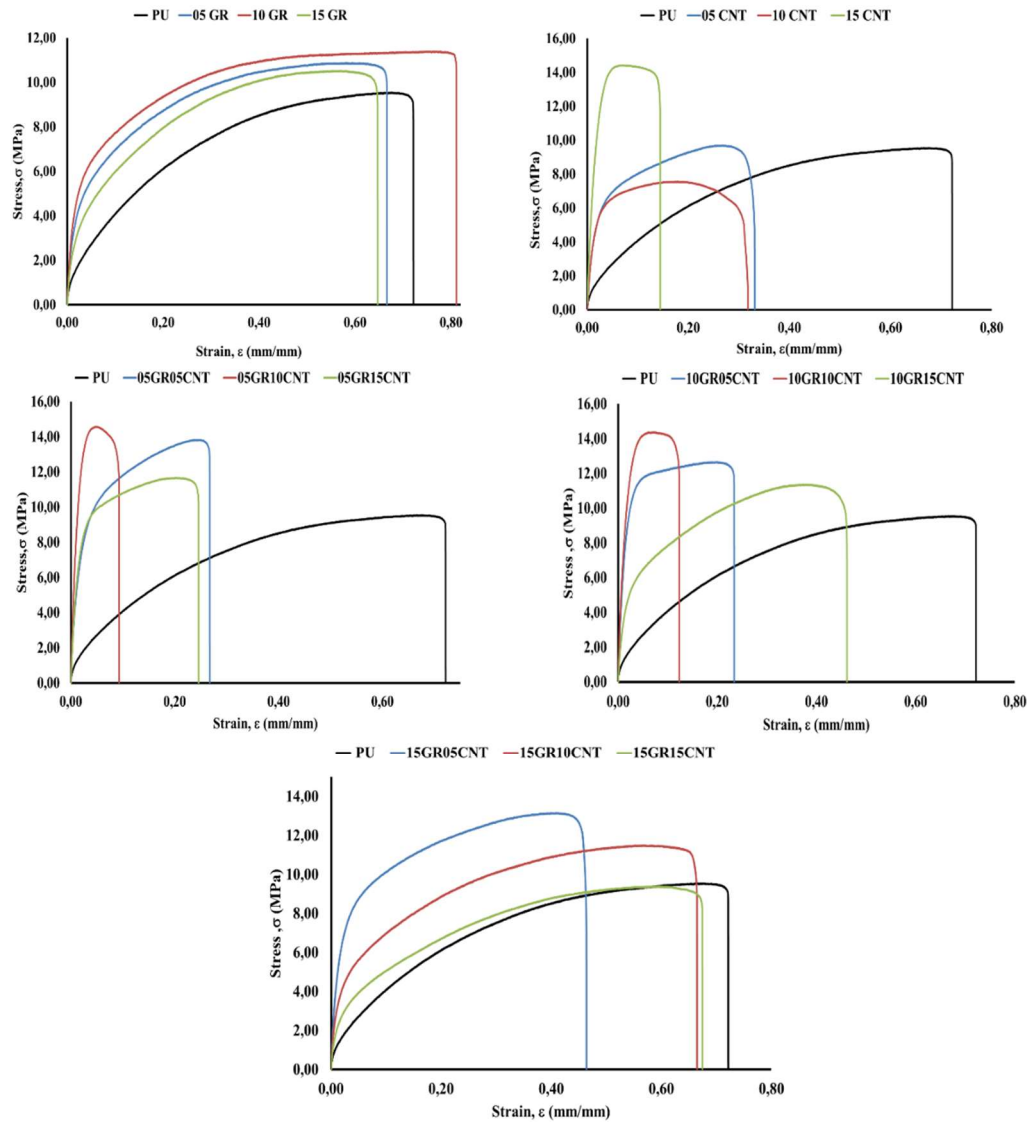


Figure 2. Stress-strain curves of pure Polyurethane adhesive and GNPs and MWCNTs nanoparticle doped adhesives

Table 2. Mechanical properties of pure polyurethane and graphene doped nanocomposites

Sample	Tensile Strength (MPa)	Maximum Strain (mm/mm)	Percentage Change (%)	Toughness (kJ/m ³)
PU	9.55±0.76	0.72±0.15	-	5.22±1.61
05GR	10.88±0.46	0.67±0.10	13.93	6.12±1.06
10GR	11.41±0.39	0.81±0.04	19.48	7.11±0.47
15GR	10.52±0.39	0.65±0.10	10.16	5.51±1.18

Table 3. Mechanical properties of polyurethane and MWCNT doped nanocomposites

Sample	Tensile Strength (MPa)	Maximum strain (mm/mm)	Percentage Change (%)	Toughness (kJ/m ³)
PU	9.55±0.76	0.72±0.15	-	5.22±1.61
05CNT	9.71±0.09	0.33±0.08	1.68	2.71±1.02
10CNT	7.57	0.31	-20.73	1.88
15CNT	14.43	0.14	51.10	2.06

Table 4. Mechanical Properties of graphene and mwcnt doped nanocomposites

Sample	Tensile Strength (MPa)	Maximum Strain (mm/mm)	Percentage Change (%)	Toughness (kJ/m ³)
PU	9.55±0.76	0.72±0.15	-	5.22±1.61
05GR05CNT	13.83±0.95	0.27±0.05	44.82	3.07±0.49
05GR10CNT	14.58±1.49	0.09±0.012	52.67	1.17±0.19
05GR15CNT	11.67±0.56	0.25±0.08	22.20	2.56±0.97
10GR05CNT	12.66±0.99	0.23±0.04	32.57	2.71±0.57
10GR10CNT	14.37±1.34	0.12±0.03	50.47	1.66±0.58
10GR15CNT	10.57±0.52	0.46±0.05	10.68	4.02±0.54
15GR05CNT	13.15±0.59	0.46±0.07	37.70	5.42±0.79
15GR10CNT	11.51±0.84	0.66±0.09	20.52	6.31±0.98
15GR15CNT	9.71±0.91	0.67±0.12	3.98	5.33±0.67

3.2. Shear tests of single lap joints

Based on the results obtained from the dogbone tensile tests, adhesives with the best mechanical properties were selected for lap shear tensile tests and the stress-strain (shear strain) curves were plotted. These curves are shown in Figure 3. The shear strengths, shear strain values, and shear modulus derived from the stress-strain curves are seen in Table 5. The results indicate that the addition of nanoparticles led to an increase in shear strengths. The maximum shear stress was achieved with the 10GR10CNTSLJ sample as 15.51 MPa, showing a 14.80% improvement compared to the unmodified PUSLJ sample. Additionally, the 10GRSLJ sample exhibited a 13.18% increase in shear stress, and the addition of 0.10 wt.% MWCNT to the 10GR10CNTSLJ sample resulted in a noticeable improvement in shear strength. This enhancement is attributed to the higher elastic modulus of MWCNTs compared to graphene nano particles, which provides better stress transfer and joint strength [20]. The highest shear strain for the maximum shear strength was observed for the 15GR05CNTSLJ sample, which has the maximum graphene nanoparticle content and the minimum MWCNT nanoparticle content.

Table 5. Mechanical properties of graphene and MWCNT doped single lap joints

Sample	Shear Strength (MPa)	Strain For Maximum Shear Stress (mm/mm)	Percentage Change (%)	Shear Modulus (GPa)
PUSLJ	13.51±0.43	1.83±0.20		9.32±1.83
10GRSLJ	15.29±1.16	1.43±0.32	13.18	10.06±1.4
15CNTSLJ	14.99±1.20	0.86±0.42	10.95	9.43±3.47
05GR10CNTSLJ	14.17±0.82	1.15±0.40	4.89	13.50±1.2
10GR10CNTSLJ	15.51±1.72	0.98±0.56	14.80	12.91±4.2
15GR05CNTSLJ	15.11±2.07	2.05±0.63	11.84	9.81±2.07

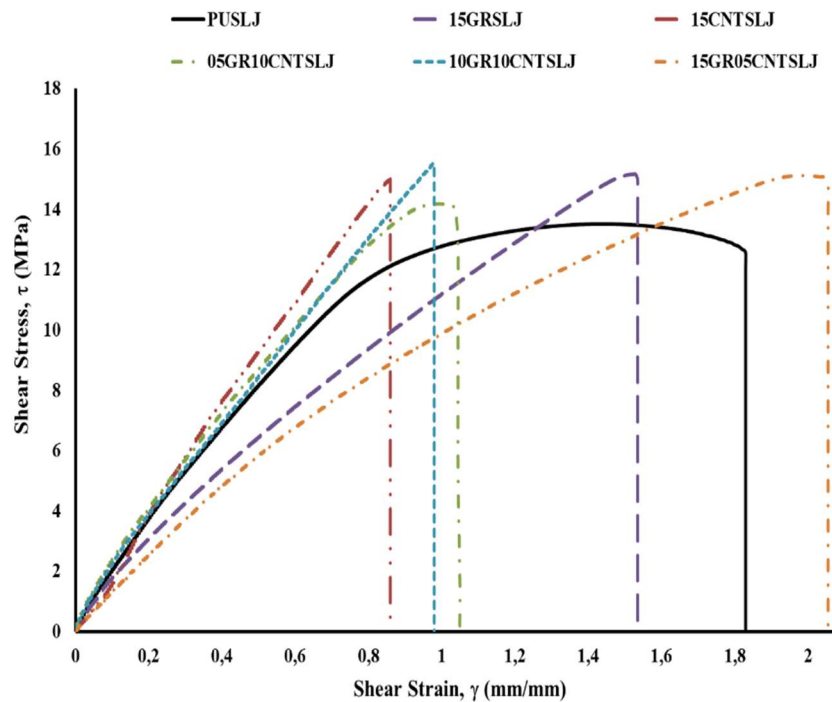


Figure 3. Shear stress- shear strain curves of single lap joints

3.3. SEM analysis of fractured surfaces of nanocomposites

After dogbone tensile and lap shear tensile tests, fracture surfaces were examined by ZEISS Evo LS 10 Scanning Electron Microscope (SEM). The fracture surfaces provide initial insights into the effects of nanoparticle additives on fracture behavior and mechanisms [21]. As shown in Figure 4a, the examination of neat polyurethane sample fracture surfaces reveals a smoother and more uniform surface, indicative of the material's weak resistance to crack initiation and propagation, characteristic of a brittle fracture process. After the tensile test, the fracture surfaces of thermosetting polymers display a mirror-like region, where cracks initially propagate slowly and then suddenly accelerate, forming a relatively smooth area. This mirror region is followed by a more pronounced rough zone with significant surface irregularities and flow lines (river pattern), which forms as the deformation rate increases, reflecting the final crack propagation [22]. Polymer-based adhesives can be considered more brittle than nanoparticle-reinforced adhesives due to the density of mirror regions, which indicates a weaker resistance to crack initiation and propagation [23]. The addition of nanoparticles to polyurethane adhesives enhances mechanical properties within the composite structure through various toughening mechanisms. These include nanoparticle rupture, crack development, bridging, shear band formation, plastic deformation, crack pinning, and crack bending [24]. SEM images of the 15GR tensile sample are presented in Figure 4b that highlights the regions of embedded graphene and matrix fracture on the fracture surface. Comparing these images reveals that the surface has a rougher texture and that the graphene effectively inhibits crack propagation [25]. In Figure 4b, the single-lap joint sample exhibits coarser fracture surfaces and features steeply inclined structures on its surface. Typically, increased surface roughness accompanies plastic deformation of the matrix, resulting in higher fracture energy expenditure. Consequently, the improved dispersion and distribution of graphene within the adhesive matrix led to enhanced shear strength and toughness values due to increased energy distribution during the fracture process. These indicators suggest cohesive damage in the adhesive region, thus confirming the high quality of the prepared composite surfaces and the produced nanocomposite adhesive [26].

SEM images of the 15CNT tensile sample are presented in Figure 4c, while SEM images of the 15CNTSLJ sample are shown in Figure 5c. The images clearly illustrate that the MWCNTs effectively arrest crack propagation across the matrix fracture surfaces through a bridging mechanism. The SEM images of the 15GR05CNT SLJ sample (Figure 5d) reveal that the addition of MWCNTs results in rougher regions on the

fracture surfaces of the adhesives compared to pure polyurethane. Significant toughness increasing observed in MWCNT-reinforced polyurethane adhesives include nanotube pull out, delamination, and bridging effects, which contribute to changes in crack propagation direction or branching[27]. In the 15GR05CNTSLJ sample (Figure 5d), the surface seems an even rougher texture due to the combined addition of graphene and MWCNTs. The nanoparticle reinforcement effectively inhibits crack propagation, thereby enhancing the toughness of the adhesive.

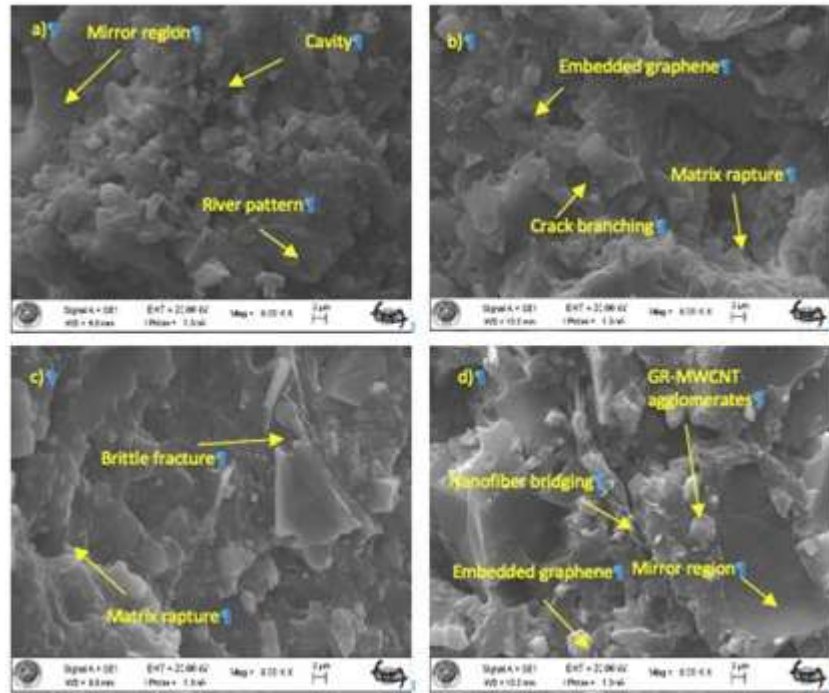


Figure 4. SEM images of nanocomposite materials' fractured surfaces of doped with MWCNT and graphene after tensile tests at 10kX magnification a) Neat polyurethane, b) 0.15% graphene doped c) 0.15%MWCNT doped, d) 0.10% graphene and 0.10%MWCNT doped

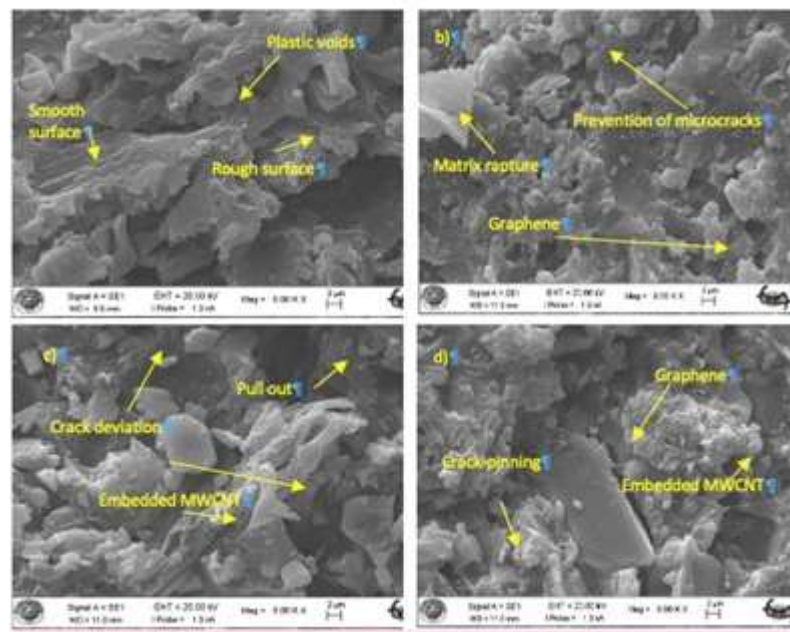


Figure 5. SEM images of nanocomposite materials' fractured surfaces of doped with MWCNT and graphene after shear tests at 10kX magnification a) Neat polyurethane, b) 0.15% graphene doped c) 0.15%MWCNT doped, d) 0.15% graphene and 0.05%MWCNT doped

3.3. Fractured surfaces of nanocomposites

Figure 6 presents the fracture surfaces of samples exhibiting the best mechanical properties resulting from shear strength tests of SLJs. There is no significant difference observed in the fracture surfaces among different nanoparticle-reinforced adhesives in terms of mechanical properties. The appearance of damaged surfaces in almost all SLJ samples is approximately the same. It is observed that the damage mechanisms of SLJ samples are cohesive failure because of adhesive remained on both fracture surfaces [28]. Examining the fracture surfaces of PUSLJ, 15GRSLJ, and 15CNTSLJ samples, no air cavities were observed. However, air cavities were observed in samples with high nanoparticle contents, such as 05GR10CNTSLJ, 10GR10CNTSLJ, and 15GR05CNTSLJ. This condition is interpreted as contributing to the reduction in mechanical properties [29].

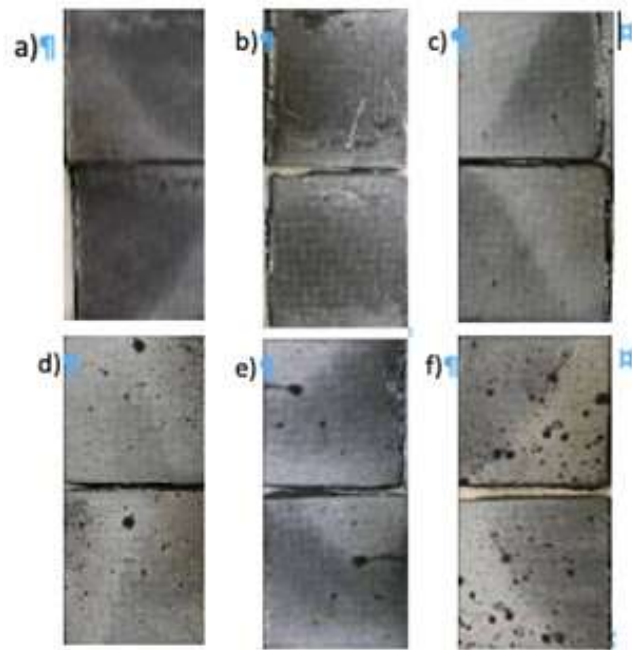


Figure 6. Macroscopic images of the broken surfaces of SLJ connections;
a) PUSLJ, b) 15GRSLJ, c) 15CNTSLJ, d) 05GR10CNTSLJ e) 10GR10CNTSLJ and f) 15GR05CNTSLJ

3. Results and Discussion

In this study, the mechanical properties of polyurethane nanocomposite adhesives, reinforced with graphene and MWCNT nanoparticles were experimentally investigated. For this purpose, SLJ and dogbone tensile samples were prepared, and their mechanical properties were examined. The major conclusions are presented as follows:

When examining the experimental results,

- It is observed that the addition of graphene nanoparticles increases the tensile strength of the samples while causing a decrease in the maximum strain values at failure. Additionally, the increasing in toughness values was observed with the increase in graphene content. If the MWCNT were added to the polyurethane adhesive, an increase in tensile strength was observed, while a decrease in unit strain values occurred. In contrast to the addition of graphene nanoparticles, the incorporation of MWCNT nanoparticles resulted in a decrease in toughness values.

- When graphene and MWCNT nanoparticles were used together as additives in dogbone tensile samples, the highest values for tensile strength, maximum strain, and toughness were achieved when the graphene content was maximum and the MWCNT content was minimum.

- Improvement in mechanical properties was observed compared to the pure polyurethane adhesive when nanoparticles were added to the polyurethane adhesive.

When examining the experimental results of composite-to-composite SLJ samples,

- An increase in shear strength was observed in all SLJs. The highest shear strength was observed in sample 10GR10CNTSLJ, where the graphene nanoparticle content was maximum and the MWCNT nanoparticle content was minimum. Upon examining the strain values at the maximum shear strength, it was observed that the maximum strain values decreased with the addition of MWCNT nanoparticles, while an increase in strain values was observed with the increase in graphene nanoparticle content.

- In SEM images, it has been determined that MWCNT nanoparticles and graphene nanoparticles halt the propagation of cracks between fracture surfaces by bridging, or causing the crack to branch. The formation of these types of damage indicates that a homogeneous mixture was achieved and that nanoparticle reinforcement, aimed at enhancing the strength and toughness of the adhesive, was successful.

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Conflict of Interest Statement

The authors declare that there is no conflict of interest.

References

- [1] ASTM D907, "Standard Terminology of Adhesives," *American Society for Testing and Materials*, 2011. doi: 10.1520/D0907-12A.2
- [2] A. Y. Kanani, X. Hou, and J. Ye, "A novel dissimilar single-lap joint with interfacial stiffness improvement," *Composite Structures*, vol. 252, Nov. 2020. doi: 10.1016/J.COMPSTRUCT.2020.112741
- [3] A. J. Kinloch, "Adhesion and Adhesives," Dordrecht: Springer Netherlands, 1987. doi: 10.1007/978-94-015-7764-9
- [4] H. Dodiuk, I. Belinski, A. Dotan, and S. Kenig, "Polyurethane adhesives containing functionalized nanoclay," *Journal of Adhesion Science and Technology*, vol. 20, no. 12, pp. 1345–1355, 2006. doi: 10.1163/156856106778456573
- [5] P. Galvez, J. Abenojar, and M. A. Martinez, "Durability of steel-CFRP structural adhesive joints with polyurethane adhesives," *Composites Part B: Engineering*, vol. 165, pp. 1–9, May 2019. doi: 10.1016/j.compositesb.2018.11.097
- [6] N. Naat, Y. Boutar, S. Naïmi, S. Mezlini, and L. F. M. Da Silva, "Effect of surface texture on the mechanical performance of bonded joints: a review," *The Journal of Adhesion*, vol. 99, no. 2, pp. 166–258, Jan. 2023. doi: 10.1080/00218464.2021.2008370
- [7] J. M. Wernik and S. A. Meguid, "On the mechanical characterization of carbon nanotube reinforced epoxy adhesives," *Materials and Design*, vol. 59, pp. 19–32, 2014. doi: 10.1016/j.matdes.2014.02.034
- [8] C. Guo *et al.*, "Mechanical and thermal properties of multiwalled carbon-nanotube-reinforced Al₂O₃ nanocomposites," *Ceramics International*, 2020. doi: 10.1016/j.ceramint.2020.04.039
- [9] H. Ejaz, A. Mubashar, E. Uddin, Z. Ali, and N. Arif, "Effect of functionalised and non-functionalised GNPs addition on strength properties of high viscous epoxy adhesive and lap shear joints," *Polymer Testing*, vol. 113, p. 107680, 2022. doi: 10.1016/j.polymertesting.2022.107680
- [10] M. De Volder, S. Tawfik, R. Baughman, and A. J. Hart, "Carbon Nanotubes: Present and Future Commercial Application," *Science*, vol. 339, pp. 535–539, 2013. doi: 10.1126/science.1222453

- [11] G. Otorgust, H. Dodiuk, S. Kenig, and R. Tenne, "Important insights into polyurethane nanocomposite-adhesives; a comparative study between INT-WS2 and CNT," *European Polymer Journal*, vol. 89, pp. 281–300, Apr. 2017. doi: 10.1016/j.EURPOLYMJ.2017.02.027
- [12] Ö. Özbek and M. Çakır, "MWCNT and Nano-silica Hybrids Effect on Mechanical and Fracture Characterization of Single Lap Joints of GFRP plates," *International Journal of Adhesion and Adhesives*, vol. 117, p. 103159, 2022. doi: 10.1016/j.ijadhadh.2022.103159
- [13] Z. Jia and G. Yuan, "Numerical study on the mechanical behavior of a polyurethane adhesive under high strain rate," *Composites Part B: Engineering*, vol. 158, pp. 131–140, 2019. doi: 10.1016/j.compositesb.2018.08.110
- [14] Z. Jia, J. Yu, Q. Liu, S. Yu, and Z. Wang, "Functionally graded adhesive joints with exceptional strength and toughness by graphene nanoplatelets reinforced epoxy adhesives," *International Journal of Adhesion and Adhesives*, vol. 125, p. 103402, Jul. 2023. doi: 10.1016/j.ijadhadh.2023.103402
- [15] H. Ejaz, A. Mubashar, E. Uddin, Z. Ali, and N. Arif, "Influence of MWCNTs on Strength Properties of High Viscous Epoxy Adhesive and Fracture Behavior of Adhesively Bonded Joints," *Theoretical and Applied Fracture Mechanics*, vol. 120, p. 103412, 2022. doi: 10.1016/j.tafmec.2022.103412
- [16] H. Ejaz, A. Mubashar, M. A. M, and S. Waqar, "Effect of GNP and MWCNT Addition on Lap Shear Strength of Adhesively Bonded Joints," in *2022 19th International Bhurban Conference on Applied Sciences and Technology (IBCAST)*, IEEE, Aug. 2022. pp. 116–121. doi: 10.1109/IBCAST54850.2022.9990107
- [17] A. Hallal, A. Elmarakbi, A. Shaito, and H. El-Hage, "Overview of Composite Materials and their Automotive Applications," *Advanced Composite Materials for Automotive Applications: Structural Integrity and Crashworthiness*, pp. 1–28, 2013. doi: 10.1002/9781118535288.CH1
- [18] ASTM D638, "Standard Test Method for Tensile Properties of Plastic," Available: <https://www.astm.org/d0638-10.html>. [Accessed: Oct. 09, 2022].
- [19] ASTM D1002-10, "Standard Test Method for Apparent Shear Strength of Single-Lap-Joint Adhesively Bonded Metal Specimens by Tension Loading (Metal-to-Metal)," in *ASTM Book of Standards*, ASTM International, 2019. pp. 1–5. doi: 10.1520/D1002-10R19
- [20] M. V. Cakir and D. Kinay, "MWCNT, nano-silica, and nano-clay additives effects on adhesion performance of dissimilar materials bonded joints," *Polymer Composites*, vol. 42, no. 11, pp. 5880–5892, Nov. 2021. doi: 10.1002/pc.26268
- [21] X. Jia, B. Liu, L. Huang, D. Hui, and X. Yang, "Numerical analysis of synergistic reinforcing effect of silica nanoparticle–MWCNT hybrid on epoxy-based composites," *Composites Part B: Engineering*, vol. 54, pp. 133–137, Nov. 2013. doi: 10.1016/j.compositesb.2013.04.002
- [22] M. S. Goyat, S. Suresh, S. Bahl, S. Halder, and P. K. Ghosh, "Thermomechanical response and toughening mechanisms of a carbon nano bead reinforced epoxy composite," *Materials Chemistry and Physics*, vol. 166, pp. 144–152, 2015. doi: 10.1016/j.matchemphys.2015.09.038
- [23] M. Rashad *et al.*, "Effect of graphene nanoplatelets (GNPs) addition on strength and ductility of magnesium-titanium alloys," *Journal of Magnesium and Alloys*, vol. 1, no. 3, pp. 242–248, 2013. doi: 10.1016/j.jma.2013.09.004
- [24] F. Ghadami, M. R. Dadfar, and M. Kazazi, "Hot-cured epoxy-nanoparticulate-filled nanocomposites: Fracture toughness behavior," *Engineering Fracture Mechanics*, vol. 162, pp. 193–200, 2016. doi: 10.1016/j.engfracmech.2016.05.016
- [25] R. Rodríguez, B. Pérez, and S. Flórez, "Effect of different nanoparticles on mechanical properties and curing behavior of thermoset polyurethane adhesives," in *Journal of Adhesion*, Taylor and Francis Inc., 2014. pp. 848–859. doi: 10.1080/00218464.2014.893509
- [26] Y. Wei, X. Jin, Q. Luo, Q. Li, and G. Sun, "Adhesively bonded joints – A review on design, manufacturing, experiments, modeling and challenges," *Composites Part B: Engineering*, vol. 276, p. 111225, May 2024. doi: 10.1016/j.compositesb.2024.111225
- [27] W.-S. Kim, I.-H. Yun, J.-J. Lee, and H.-T. Jung, "Evaluation of mechanical interlock effect on adhesion strength of polymer–metal interfaces using micro-patterned surface topography," *International Journal of Adhesion and Adhesives*, vol. 30, no. 6, pp. 408–417, Sep. 2010. doi: 10.1016/j.ijadhadh.2010.05.004
- [28] A. Nemati Giv, M. R. Ayatollahi, S. H. Ghaffari, and L. F. M. da Silva, "Effect of reinforcements at different scales on mechanical properties of epoxy adhesives and adhesive joints: a review," *The Journal of Adhesion*, vol. 94, no. 13, pp. 1082–1121, Nov. 2018. doi: 10.1080/00218464.2018.1452736

[29] R. Moriche, M. Sánchez, A. Jiménez-Suárez, S. G. Prolongo, and A. Ureña, "Strain monitoring mechanisms of sensors based on the addition of graphene nanoplatelets into an epoxy matrix," *Composites Science and Technology*, vol. 123, pp. 65–70, Feb. 2016. doi: 10.1016/j.compscitech.2015.12.002

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