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## **The Effect of Nano Graphene Reinforcement on Pin and Adhesively Bonded Sandwich Composite Structures**

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**Abstract:** The subject of joining composite parts is an important issue in a wide range of engineering applications. Currently mechanically-fastened and adhesively bonded joints are the two established and accepted techniques of joining used in aerospace industry for assembling composite structures. More effective adhesively bonded joints can be achieved by increasing the chemical properties of the adhesive. In this paper, nano graphene particles were added to adhesive in various proportions (0.5% wt., 1% wt. and 1.5% wt.) to increase the mechanical strength of the pin and adhesively bonded (hybrid, bolted/bonded) sandwich composite structures. The mechanical properties of the newly produced nano graphene added adhesives were determined experimentally. Sandwich composite structure hybrid models with holes were created using finite elements method. The two dimensional plane strain and three dimensional analyses have been carried out. Effect of the nano graphene reinforced and hybrid structures on the all displacements were discussed on tensile property of the sandwich structures. The sensitivity of mechanical response to the compressibility of the adhesive material has been demonstrated. Numerical analysis of nano graphene reinforced hybrid joints show their strength life is longer than corresponding adhesively bonded/bolted joints. Furthermore, hybrid joints can modify the tension-compression area distribution at the side of the bolt hole, thus making composite material participate more in the load bearing. This is reflected in the increased bolt hole strain and better structural performance.

**Keywords:** Composites, Hybrid joints, Nano graphene reinforced adhesive, Finite element analysis

### **Introduction**

For many years, designers have been able to replace parts made from common materials such as steel and composite materials. The most obvious benefit of composite materials is that they can be designed to achieve certain material properties in the desired direction. Until now, thin composite plates are widely used in different industries. For this reason, design and analysis methods for thin composites have been widely developed (Reddy, 1997). and various techniques have been invented by manufacturers to produce more efficient and complex parts and several techniques have been invented by manufacturers to produce more complicated parts (Van Hoa, 2009).

For efficient connection designs, it is necessary to take advantage of all the properties of composite materials, which are increasingly used in aircraft structures. Some of the applications of bolt joints in aerospace industry are such as wing to fuselage joint in Boeing/MDD Harrier, Boeing/Bell V-22 Osprey, Boeing 777 and Grumman X-29 (NASA) (Heslehurst, 2013). Bolted joints, the formerly and currently preferred method for joining these structures, suffer from low joining efficiency, resulting in thicker structures in the joint region. Typically, a composite joint has been found to provide a joint efficiency of 40-50% compared to 60% for metals. (Bodjona et al., 2015).

The wing of an Airbus 380 alone consists of more than 30,000 elements, with approximately 750,000 bolted connections (Zhang & Zhao, 2012). These joints are of key importance as they constitute the weakest point that causes the element to break. The three main methods of joining composite laminates together are either

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mechanical fastening, bonding, or a combination of the two, called "hybrid" bonding. Mechanical fasteners such as pins, rivets and bolts have been known to occupy a very large area in the aerospace industry for decades (Ireman et al., 1993; Khudhayer & Othman, 2011; Collings, 1997; Wei et al., 2013). The main problem with mechanical joints is the high stress concentrations that occur around the fastener holes and cause damage and this is more severe in composite laminates than in metal plates (Hart-Smith, 1998).

Adhesive bonded joints are structurally more useful and efficient than mechanically bonded joints, as they perform better in distributing loads and eliminate many of the problems of high stress concentrations associated with bolted connections (Hart-Smith, 1982 & Hart-Smith, 1985). Sandwich structures are also joined by film adhesive between layers. The bonded material is used to bear compressive loads and axial tension, while the adhesive is used to carry shear loads and provide support against compressive loads normal to the bonded ones. The materials used in the construction of sandwich structures can vary depending on the application. The adhesives of a sandwich structure are typically made of aluminum alloys, titanium, high tensile steel or multilayer ply composites, functionally graded materials (Kassapoglou, 2010; Paik et al., 1999; Uysal & Guven, 2015).

In this presented study, the plates made of composite material were connected to each other by both adhesive and bolted connection (hybrid connection), but with a big difference, in the new presented study, the bond strength was tried to be increased by adding nano graphene particles into the adhesive. Researchers carry out various studies in order to increase the mechanical properties and bond strength of adhesive joints. Some of these studies include the addition of powders such as copper powder, aluminum powder, calcium silicate, calcium carbonate, silicon oxide, titanium oxide, carbon nanotube to the epoxy adhesive. impact resistance, abrasion resistance, etc. carried out to improve mechanical properties (Wetzel et al., 2003; Meguid & Sun, 2004; Kilik & Davies, 1989; Zhai et al., 2006; Wetzel, 2006; Xian et al., 2006; Zhai et al., 200; Zhao et al., 2008; Gerson et al., 2010).

In these studies, it was stated that the particle reinforcement gave better results than the additive-free epoxy adhesive. In the finite element study, in which the effect of micro aluminum powder added to epoxy adhesive on mechanical properties was examined, it was stated that the addition of micro aluminum powder increased the bond strength (Kahraman, 2008). When multi-walled carbon nanotubes are added to the epoxy adhesive as reinforcement, the mechanical strength increases with 0.5% carbon nanotube addition. while it deteriorates when 1% and 2% carbon nanotubes are added (Pilawka et al., 2012). In another study, it was stated that when the carbon nanotube addition is around 1.5%, the mechanical properties increase and after this value, the mechanical properties decrease and even lower results than the additive-free epoxy adhesive (Wernik & Meguid, 2014). The adhesive obtained with silicate nanoparticles added to the epoxy resin at the rates of 0.5%, 1%, 1.5% and 2% by weight was used in aluminum joints to provide light and high strength. The effects of silicate nanoparticles were observed in peeling and shear stresses (Charitidis, 2020). In experimental studies using two different nanoparticles, single lap and hybrid bonding joints were investigated. Appropriate nanoparticle selection was made in experimental studies, which showed an improvement of approximately 10% in tensile strength. Adhesion zones were observed by SEM (Scanning Electron Microscopy) analyzes and the distribution of nanoparticles was investigated (Borghesi et al., 2019).

Connections where adhesive zone defects are due to environmental factors and galvanized steel materials are bonded with nanoparticle added adhesive were examined. Experimental studies were carried out for nanoparticles added into the adhesive zone at different temperatures and residence times, and their shear strength and tensile strength were investigated (Nascimento et al., 2021). In the study, in which nanoparticle reinforced adhesive is obtained and the distribution of these particles in the adhesive is primarily homogeneous and then functionally graded, the obtained adhesive properties are examined. The strength and local plastic deformations of the joints bonded with the obtained adhesive were observed (Jojibabu et al., 2020). In this study, in which both iron oxide particles and nanoparticles were added to improve the properties of thermoplastic adhesives, additions were made at the rates of 0.1%, 0.5% and 1% by weight. Single lap joints were adhered with these new adhesives and the loading displacement curves were obtained and interpreted as a result of the experiment (Ciardiello et al., 2021).

As can be seen from the studies on increasing the mechanical properties of adhesive joints and adhesives, it is understood that various powders are used. In addition, it is stated in the studies that the additive ratio is very important especially in the addition of carbon-based powders to the epoxy adhesive and affects the mechanical properties of the adhesive joint. For this reason, sandwich joint models were created with nano graphene reinforced adhesives at various rates within the scope of this study. In this study, the strengthening parameters in the adhesive region were well analyzed. The aim of this study is to examine the stress and strain behaviors on

hybrid composite sandwich structures bonded using nano graphene added adhesives and to evaluate the effect of nano graphene additive on bond strength.

## Materials and Method

### Preparation of Nano Graphene Added Epoxy Based Nanocomposite Adhesives

Nanocomposite adhesives were produced by adding 0.5%, 1% and 1.5% by weight nano graphene to the epoxy adhesive. Before preparing the nanocomposite adhesives, the nano graphene particles were dried in an oven at 120°C for 1 hour to remove moisture. While preparing the epoxy adhesive, first of all, the epoxy adhesive was weighed on a precision scale, and then the nano particles, which were weighed in a precision balance at a predetermined rate, were added to this plain epoxy adhesive. In addition, PVP (Polyvinylpyrrolidone) was added to the mixture in an equal amount with the weight of the nano particles in order to ensure homogeneous dispersion of carbon-based nano particles in the epoxy matrix. The material properties of the adhesive and nano graphene used are given in the Table 1 and Table 2.

Table 1. Mechanical properties of Loctite® EA9492 adhesive

Properties	Loctite-Hysol 9464
Shear Strength (MPa)	20
Peel Strength (MPa)	10.6
Modulus of Elasticity, E (GPa)	1.65
Shear Modulus, G (GPa)	0.75
Poisson's ratio, $\nu$	0.356

Table 2. Properties of nano graphene used in experiments

Properties	Nano Graphene
Average particle diameter ( $\mu\text{m}$ )	7
Modulus of Elasticity (GPa)	1000
Average particle thickness (nm)	6
Tensile Strength (GPa)	10-20
Density ( $\text{g}/\text{cm}^3$ )	~2
Surface area ( $\text{m}^2/\text{g}$ )	130

Within the scope of the presented paper, bulk samples were prepared and subjected to tensile test for the determination of mechanical properties of adhesives with 0.5%, 1% and 1.5% additives by weight. ISO 15166 standard was taken into account in the preparation of bulk samples. This standard specifies the test specimen dimensions required for the preparation of bulk specimens from two-component liquid adhesives (Figure 1).

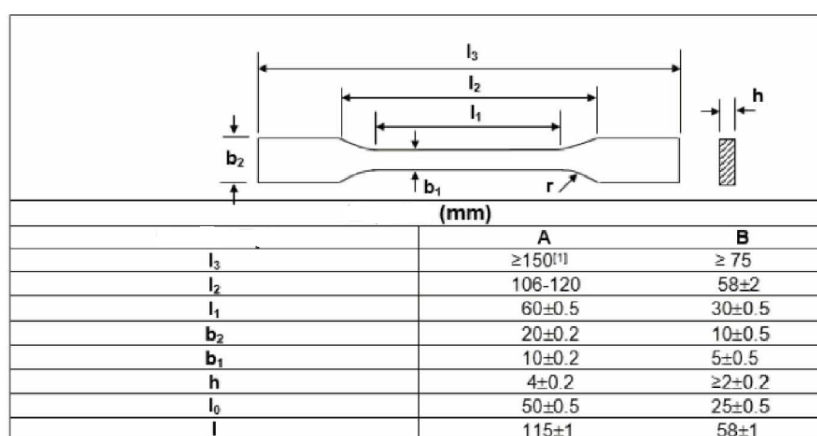


Figure 1. Tensile test sample dimensions according to ISO 15166 standard

In order to produce bulk samples, molds were made from PLA material using the additive manufacturing method (Figure 2(a)). Adhesives containing nano graphene, prepared, were poured into the mold as seen in Figure 2(b). In order to adjust the sample thickness (1.5 mm), a flat plate was closed over the mold and a weight was placed on it. As recommended in the adhesive data sheet, 72 hours were allowed at room temperature for

curing. In order for the adhesive to come off easily, one side of both the mold and the plate is covered with a 0.1 mm thick stretch film.



Figure 2. a) Mold prepared for bulk samples, b) bulk samples poured into the mold

After the prepared bulk samples were removed from the mold, they were sanded with fine sandpaper to remove the burrs. Tensile tests were first performed on the undoped one of these samples and the mechanical properties of the adhesive were compared with the adhesive data sheet. After this convergence, the mechanical properties of the adhesives with 0.5%, 1% and 1.5% nano graphene structure by weight were determined. It is planned to use the determined mechanical properties of nano graphene added adhesives in finite element analysis. Adhesive material properties, stress strain curve excel data obtained from tensile controls were entered into the ANSYS software.

### Modeling of Sandwich Composite Structures with Pin and Adhesive

Both the adherends were composite laminated plate with layup of  $[0-90]_{10s}$ . The thickness of each layer is 0.5 mm. The mechanical properties of the bonded composite material are given in the Table 3. Also, the two-hole sandwich composite plates that make up the model are shown in Figure 3(a). In Figure 3(b), the dimensions of the composite plates were given.

Table 3. Mechanical properties of composite adherends (Canyurt et al., 2010)

Properties	Composite
Modulus of Elasticity, E (GPa)	$E_{11}=22$
	$E_{22}=22$
	$E_{33}=9$
Shear Modulus, G (GPa)	$G_{12}=5.3$
	$G_{23}=3.1$
	$G_{13}=3.1$
Poisson's ratio, $\nu$	$\nu_{xy}=0.27$
	$\nu_{yz}=0.38$
	$\nu_{xz}=0.38$

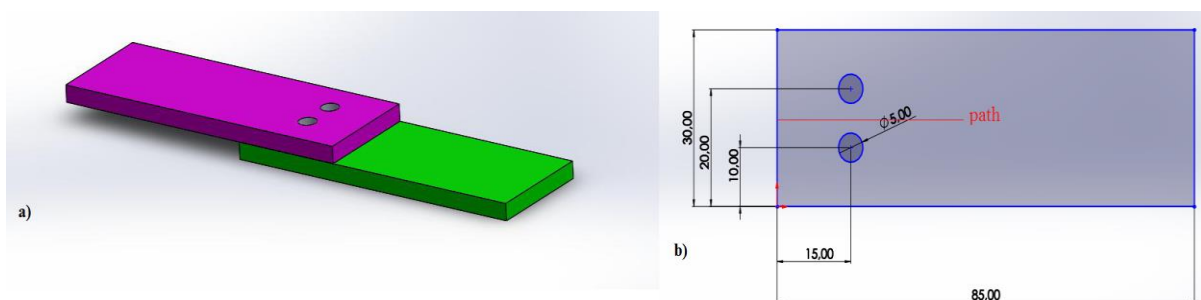


Figure 3. a) Model for composite sandwich adherend, b) Dimension of adherend

Hybrid joint models were created using the finite element method, using the bonded composite and adhesives with added 0.5%, 1% and 1.5% nano graphene, respectively. The schematic drawing of pin and adhesive bonded structure shows in Figure 4.

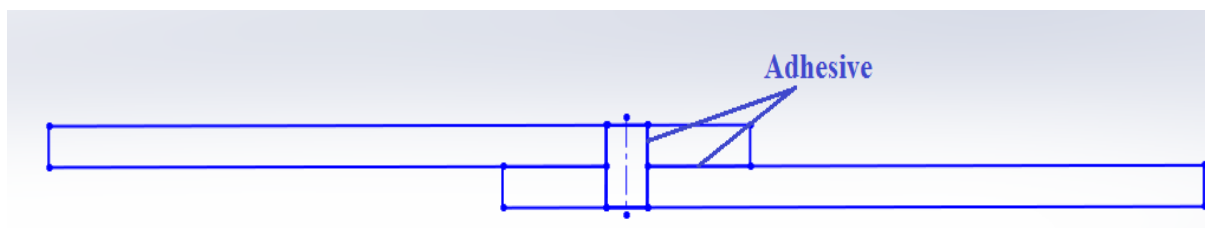


Figure 4. The schematic illustration of pin and adhesive bonded structure

### Finite Element Model

The numerical model of the fixed point supported (on holes and on plates) sandwich panel is developed by using the commercial software package ANSYS. This finite element program enables the prediction of stress and strain global behavior of sandwich panel subjected to tensile load (50 MPa) cases. The thickness of one panel is 10 mm, the adhesive thickness is 1.5 mm and the total thickness of sandwich panel,  $t = 21.5$  mm. The holes are constructed at the corners 15 cm from the edges with a diameter  $d$  of 5 cm (Figure 3). The numerical model of FGPMs sandwich panel was divided into a finite number of elements satisfying the equilibrium and compatibility at each node. The size of elements was determined for the finer mesh in the critical regions such as the holes. The mesh size was determined by the solid element constraints that the length ratio of element edges cannot be smaller than  $1/20$  and the angle between element edges cannot be less than  $70^\circ$ . Consequently, the optimal combination of mesh accuracy and elements size was found. The model has contact pairs, adhesive and adherend plates have contact position.. Surface to surface contact elements (CONTA 174 and TARGE 170) were set the overlap surface of the adherends and adhesive and pin and adhesive surfaces (Figure 5). For the details of bonding mechanics and modeling, the relevant literature (Uysal & Guven, 2015) can be consulted.

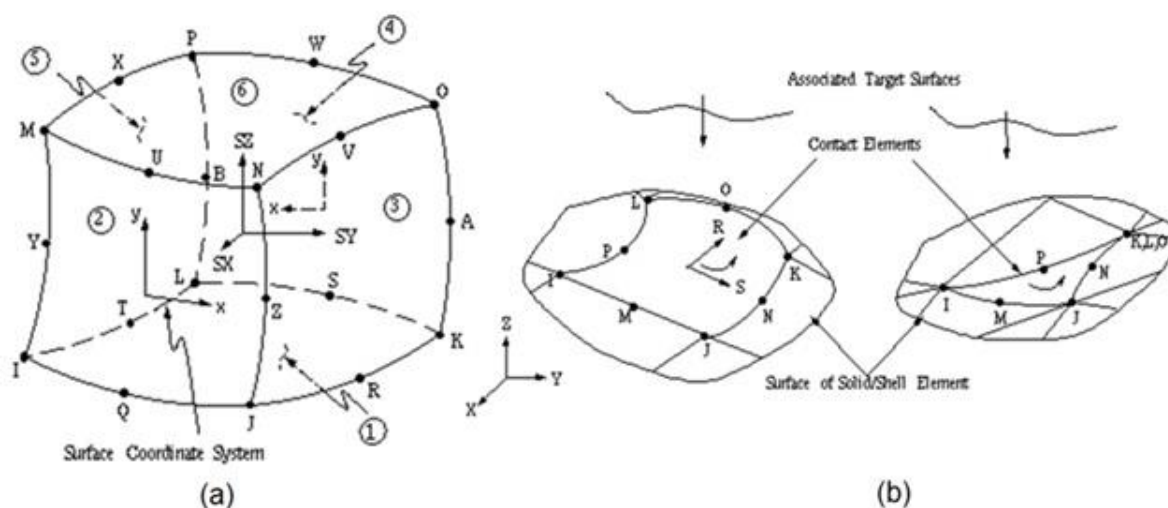


Figure 5. Element types a) Solid 95, b) Conta174 and Targe170

### Results and Discussions

Normal stresses ( $\sigma_x$  and  $\sigma_y$ ), shear stresses ( $\tau_{xy}$ ) and von mises stresses ( $\sigma_{von}$ ) were compared for all nano graphene additions (0.5% wt., 1% wt. and 1.5% wt.) in composite sandwich structures with bolt and adhesive bonds. In Figure 6 and Figure 7, stress distributions are given for hybrid joint modeled using 0.5% nanographene added adhesive. A stress distribution image of about 35 cm from the bonding area with pins is given in these. The focus is on stress distributions in the composite plate. The image with the bolt has been removed because it looks complicated.

In Figure 6(a), the  $\sigma_x$  normal stress occurring under tensile load around the hole, and in Figure 6 (b), the  $\sigma_y$  normal stress distributions occurring in this vicinity are given.

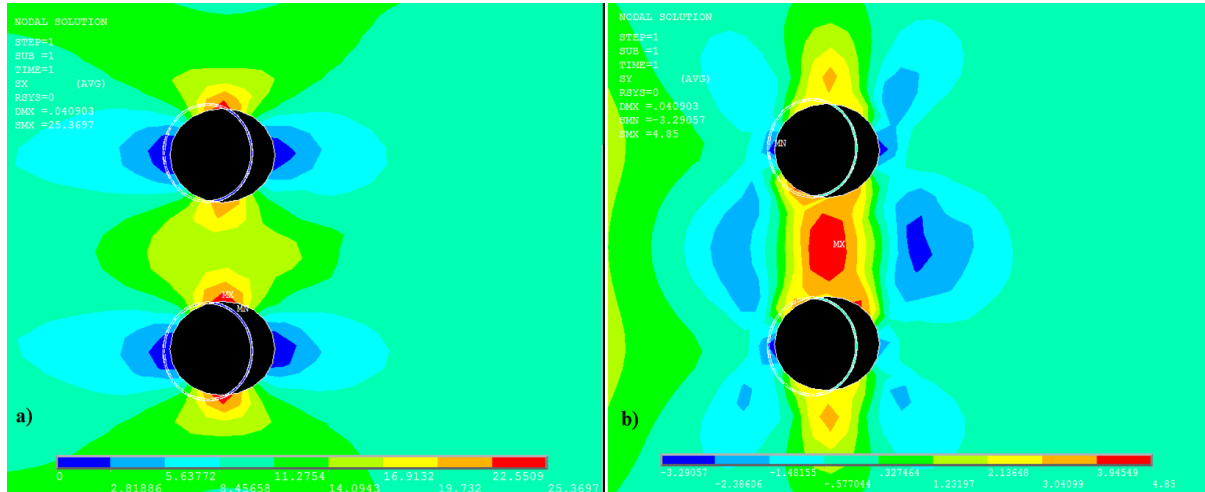


Figure 6. Stress values around the hole a) Normal stress,  $\sigma_x$ , b) Normal stress,  $\sigma_y$

The stress and strain values of the sandwiched composite plate subjected to tensile load were determined as a result of the finite element analysis. The displacement value in the X direction is maximum at the loaded edge. Its value is 4.806 mm. The maximum stress value in the X direction is 25.369 MPa. Also, the strain value in the Y direction is 4.85 Mpa. (Figure 6). Even when the loading is longitudinal, negative shear stresses occur in the composite plate. The value of these shear stresses is 5.59 MPa. The von misses stress value calculated according to the von Misses criterion is 24.32 MPa in the sandwich plate modeled with 0.5% wt. nano graphene added adhesive (Figure 7). Comparative stress plots for hybrid joints bonded by adding other weight nano graphene are given in Figure 9 and Figure 10.

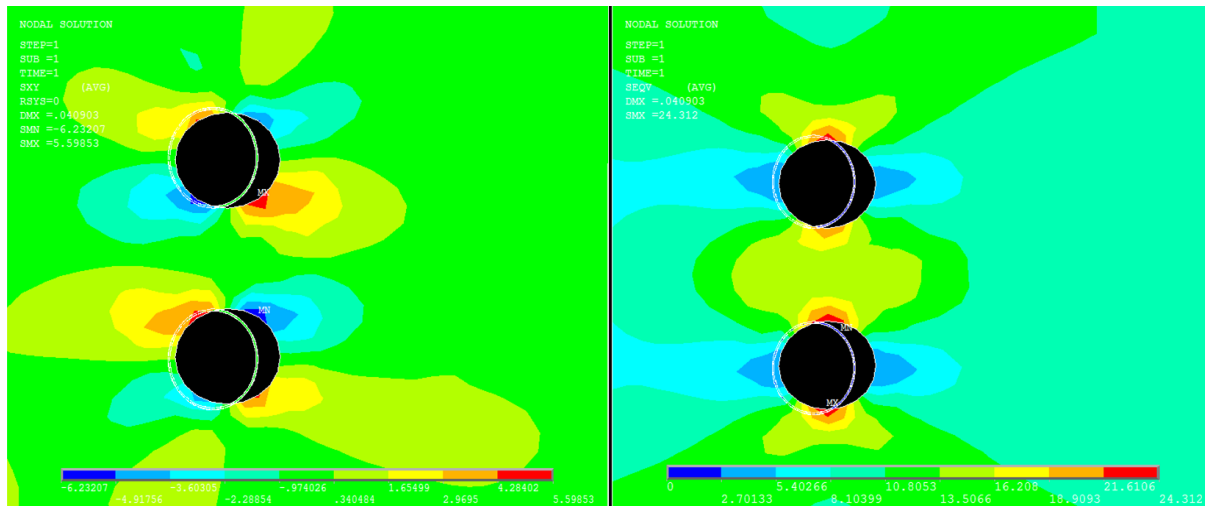


Figure 7. Stress values around the hole a) Shear Stress  $\tau_{xy}$ , b) von Misses Stress,  $\sigma_{von}$

After the stress analysis is done, a path is drawn in the finite element program for the more important regions and the stress values at the nodes there can be examined. The path is drawn for a distance of approximately 35 cm from the bonding place and in the middle of the composite plate (15 cm) (Figure 3(b)).

Normal stresses ( $\sigma_x$  and  $\sigma_y$ ), shear stresses ( $\tau_{xy}$ ) and von Misses stresses ( $\sigma_{von}$ ) behaviors are plotted on the path (Figure 8). The normal stress ( $\sigma_x$ ) value is around 8 MPa in the fixed support, this stress increased when it came to the bolted connection position and reached a value of approximately 17.45 MPa. This is proof that, in hybrid adhesives, stress concentrations are mostly on the pin side of the composite material. These stress concentrations cause separations and delaminations in the laminated composite plates, reducing the joint strength. In this study, it was aimed to improve the strength in this region by adding nano graphene particles into the adhesive. The effects of this reinforcement are given in Figure 9 and Figure 10.

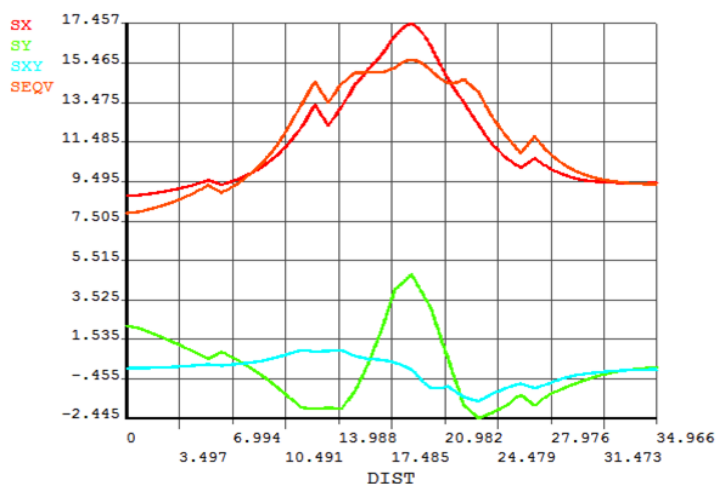


Figure 8. All stress behaviors in the adhesive bonding zone (on the composite plate)

Longitudinal tensile analysis was applied to the models to estimate the tensile strength of sandwich panels composed of composite structures.. Normal stresses ( $\sigma_x$  and  $\sigma_y$ ), shear stresses ( $\tau_{xy}$ ) and von Misses stresses ( $\sigma_{von}$ ) values were compared in order to understand the effects of nano graphene additions to the adhesive in single-layer lap joints. As can be seen in Figure 9a, the normal stress values in the x-direction are compared for an adhesive bonded sandwich panel with different nano graphene powder weight ratios (%). The normal stress values in the x direction is 25.36 MPa for the sandwich panel defined as 0.5%. This value is 20.80 MPa for a sandwich panel with 1.5% nano graphene powder added. The normal stress value in the x direction in 1% sandwich panel is between 0.5% and 1.5% and its value is 24.10 MPa.

For the composite sandwich panel with fixed support under tensile load, when the nano graphene powder weight fraction was increased from 0.5% to 1%, the normal stress in the y-direction ( $\sigma_y$ ) decreased by 6%. This decrease is 23% when the ratio is increased from 0.5% to 1.5%. The highest normal stress values were calculated for 0.5% nano graphene powder and this result showed that nano graphene powder in this ratio is more compatible with the resin and responds better to tensile load.

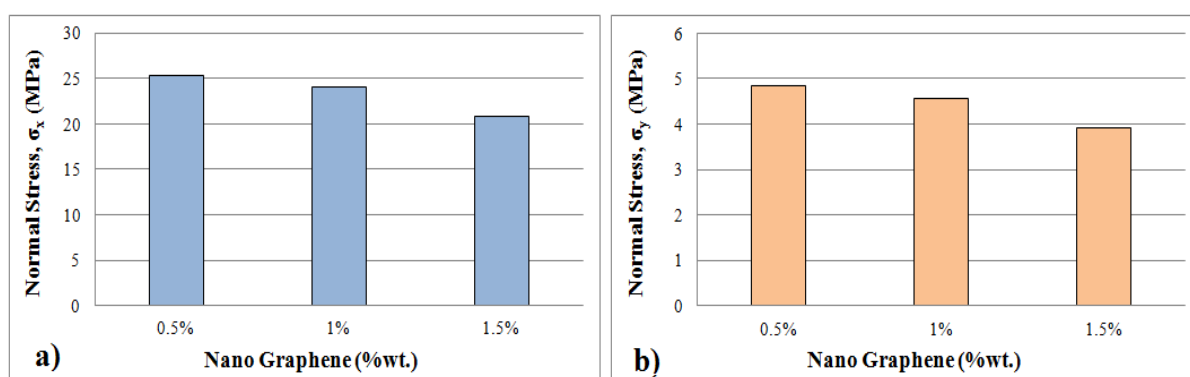


Figure 9. Effect of nano graphene weight ratio on stresses values, hybrid bond sandwich panel made of composite under tensile load, a) normal stress,  $\sigma_x$  b) normal stress,  $\sigma_y$

When considered in terms of shear stress values, the highest shear stresses are in the junction with 0.5% nano graphene powder. The formation of 5.59 MPa in the negative direction indicates that this connection has the highest probability of separation behavior caused by shear stresses. Stress values in the negative direction determine the direction of shear stress. The shear stress value of the 1% nano graphene bond is 5.11 MPa in the negative direction, which is lower than the 0.5% nano graphene bond. (Figure 10a). The results of von misses stress analysis on the composite material are important in terms of examining the joint strength and breaking behavior of the combined geometry and determining the damage to the adherend material. The occurring and von-Mises ( $\sigma_{von}$ ) stress values in Figure 10 b were determined as a result of the finite element analysis. The von Mises stress value at the hybrid bonded with 1% nano graphene added was 22.61 MPa, and this value increased by 7% with 0.5% nano graphene addition. The lowest von Mises stress value was observed in the hybrid joint bonded with 1.5% nano graphene added adhesive.

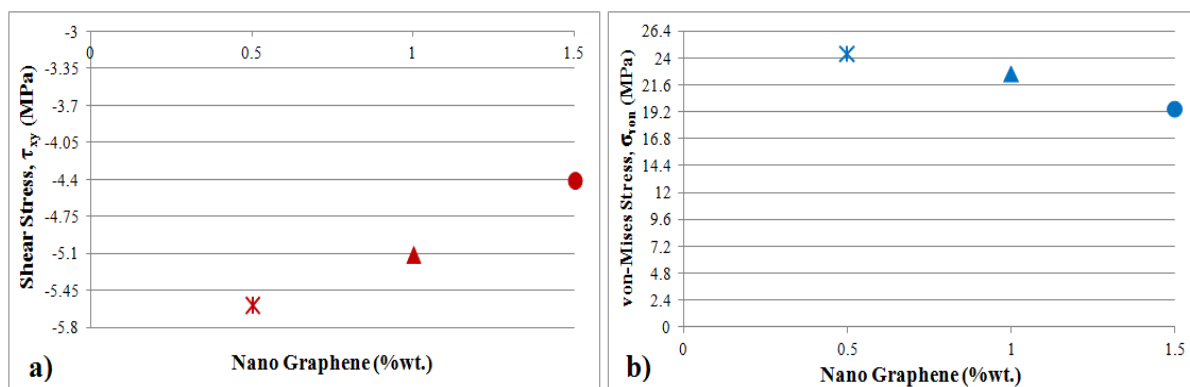


Figure 10. Effect of nano graphene weight ratio on stresses values, hybrid bond sandwich panel made of composite under tensile load, a) shear stress,  $\tau_{xy}$  b) von-misses stress,  $\sigma_{von}$

## Conclusion

In this presented study, the behavior of heterogeneous adhesive bonds is investigated by modeling them in hybrid bonds. First, the recently experimentally developed nano graphene-added adhesive model was used in adhesive materials, and secondly, the joint was modeled as a hybrid joint with both adhesive and pin. Adhesive joints made of hybrid bonds are encountered in the aerospace industry (layered connections in aircraft and shuttle hulls), land and sea transportation (at joints in car/ship skeletons) due to their superior mechanical properties, as well as their lightness and durability. Studies on adhesive bonds made of heterogeneous adhesive and layered composite with hybrid joint geometry are rare in the literature.

In this study, it was stated that when nano graphene was added to the epoxy adhesive as reinforcement, the mechanical strength increased with 0.5% nano graphene addition, while it deteriorated when 1% and 1.5% nano graphene was added. In addition, in this study, it was stated that the value of nano graphene addition has a significant effect on the bond strength in hybrid sandwich structures formed by using layered composites, and after the appropriate value, the mechanical properties of the adhesive decrease and even lower results than the pure epoxy adhesive. It is hoped that this study will be a useful guide for future research of heterogeneous bonded hybrid connections.

## Scientific Ethics Declaration

The author declares that the scientific ethical and legal responsibility of this article published in EPSTEM journal belongs to the author.

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