Failure of adhesively bonded single lap joints with MWCNT and Fullerene C$_{60}$ in the adhesive due to tensile loading

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Abstract. To create a better and stronger single lap joint that has the potential of sustaining the damage load to a greater extent, nano particles like multiwall carbon nanotube (MWCNT) and Fullerene C$_{60}$ were dispersed in the adhesive. Though the joints made by adhesive bonding are stronger in themselves but the use of nano particle can further increase the damage load carrying capacity. The current research focuses on developing adhesively bonded single lap joints (ABSLJ’s) by dispersing MWCNT and Fullerene C$_{60}$ in the adhesive at a weight percentage of 1% and 3% by ultrasonication. Carbon fiber fabric was used as adherend and a two part epoxy system was used as an adhesive. By using the metallic shims, the thickness of adhesive was retained at 0.5 mm and 1 mm. Results reveals that the joints made with the use of MWCNT showed better load bearing capacity compared to that of the joints made with Fullerene C$_{60}$. It was even revealed that with the rise in the thickness of the adhesive, the joint strength decreased. An increase of 154% in the joint strength was obtained for joint sample with 3 wt % MWCNT and 0.5 mm adhesive thickness compared to non doped joints. As the adhesive thickness was raised to 1 mm, for the same weight percentage the joint strength dipped by 16%. While for Fullerene C$_{60}$, increase in the joint strength was seen at 1 wt % with 0.5 mm adhesive thickness followed by a 15% dip in strength at 1 mm adhesive thickness.

1. Introduction
Joints are employed everywhere starting from construction to automobile, aircraft and has many other applications. With the availability of different bonding methods, these joints can be used to combine similar or dissimilar parts. One among them is adhesive bonding. The employment of adhesive bonding technique helps in providing not only a strong joint but also saves a lot of weight by avoiding the usage of nuts and bolts or rivets in creating a bond. Also the choice of adhesive for suitable application and the preparation of adherent surface are necessary before bonding [1]. Arranging the fibres in various orientations in the adherent influences the performances of the joint which can be related with the failure mechanisms [2]. Increment in the length of overlap provides better joint performance under tensile loading while for four point bending test overlap distance is not a deciding factor [3].

The introduction of nano materials in the adhesive while creating the joints can further lead to improvement in the load carrying capacity. These nano materials can be of various sizes and are of multiphase in nature [4]. Variation in the rate of loading affects the ultimate shear strength in a non linear manner when nanofillers were included in the adhesive [5]. Proper and uniform dispersion of nanofillers is necessary to avoid any lump formation. Therefore the use of proper dispersion technique...
is to be taken into consideration. Dispersion using mechanical stirrer can lead to breakage of the nanofillers [6]. Also adding the nanofillers in correct proportion can lead to increase in toughness, glass transition temperature and shear strength of the lap joint case [7]. The final displacement obtained is associated with the increase in load carrying capacity of the joints when optimum percentage of graphene nano sheets was added to the adhesive [8].

Graphene addition at 1% by weight has lead to significant enhancement in the mechanical properties. Additionally at a weight percentage of 4, it was observed that the properties deteriorate due to poor dispersion [9]. At lower percentage of single walled carbon nanotube (SWCNT) mixed in the adhesive, the peel strength and lap shear strength were not affected but with an increase in the percentage of SWCNT, peel strength was increased but a drop was observed in the lap shear strength [10]. Use of MWCNT doped adhesive produced higher joint strength in comparison to the undoped adhesive [11]. Improvement in the average shear strength was observed due to effective transfer of shear load to fibre when a suitable percent of MWCNT was added to the adhesive [12]. Also when the weight percent of MWCNT was varied from 0.1% to 0.3%, an increase in bonding strength was observed [13]. The use of aromatic amine hardener along with Fullerene nanofillers did not show any significant change in the bonding strength but the addition of Fullerene nanofillers to aliphatic amine hardener showed an increase in the bond strength [14].

Therefore, to investigate the interaction of nanofillers with the adhesive and its impact on the tensile properties of single lap joints (SLJ), nanofillers in two different weight percent was added to the epoxy adhesive. Also a variation with respect to the adhesive thickness was done to identify its effect on the joint strength. Therefore this work aims at determining the weight percentage of nanofillers and also determining the adhesive thickness for obtaining greater bond strength.

2. Experimental Work

2.1. Materials
Carbon fibre of plane weave pattern and 0/90 orientation along with a two part epoxy resin ROTEX EP 306 and hardener ROTEX EH 758 was used to prepare the laminate. The desired thickness of the adherend was obtained by using various layers of the same carbon fabric. For creating the joints a separate two part epoxy system including ROTEX EP-416 resin and ROTEX EH-153 hardener was used as adhesive. Apart from this, MWCNT of 10–15 nm average diameter, 5µm average length with 99% purity and Fullerene C60 of 99% purity were used as the nanofillers.

2.2. Laminate Preparation
Preparation of CFRP laminate as shown in Figure 1 was done by using hand layup technique. Eleven layers of carbon fabric of thickness 0.26 mm each was used for laminate preparation to obtain a desired thickness of 3 mm along with the ROTEX EP 306 resin and ROTEX EH 758 hardener mixture. The ratio of resin to hardener was maintained at 10:1 for the fabrication. A layer of resin and hardener mixture was applied on the mylar sheet and carbon fabric was placed over it. The same process was repeated till the desired thickness was obtained.

During the fabrication, the excess resin-hardener mixture was removed by the help of a roller. The laminate had waxed and cleaned mylar sheet placed on both the sides to facilitate easy removal of the fabricated laminate. The fabricated laminate was then left for curing at room temperature for 24 hours followed by curing at 60° C for 2 hours.
2.3. Specimen Preparation
Cutting of laminate as per ASTM D-5868 standard was accomplished by using controlled abrasive jet of water. The use of this cutting technique produced smooth edged specimens. The dimension of each specimen was maintained at 101.6mm * 25.4 mm as shown in Figure 2. Once the specimens were cut from the laminate, surface preparation was carried out to make the joints.

The specimens were then rubbed with 40 grit sand papers to make the bonding surface rough and then cleaned with acetone to remove any dust or loose particles. The average thickness of each specimen was measured to be 3.03 mm.

2.4. Preparation of Adhesive
The adhesive preparation for making the joints requires blending of nanofillers in the adhesive. Various methods are available for blending the nanofillers but, the nanofillers need an even and uniform dispersion in the adhesive without any coagulation and without damage to the nanofillers. Thus the method chosen here is Ultrasonication shown in Figure 3, wherein the MWCNT and Fullerene C₆₀ nanofillers in weight percent of 1% and 3% were mixed in 10gm of resin. The further steps were the same as was carried out in [13].
2.5. Joints Preparation

For the preparation of SLJ’s, the nanofillers doped adhesive was applied on the rough and clean surface of the adherend by covering the overlap distance. The top face of the adhesive was covered with the rough surface of the adherend sample giving it the form of a lap joint. To obtain uniformity in the thickness of the adhesive, suitable metal shims of 0.5 mm and 1 mm were used. The metal shims were coated with oil to facilitate easy removal after the curing of the joints. The joints made were clamped and was left for curing at room temperature. After 8 hours the metal shims were carefully removed without disturbing the alignment of the joint. Thereafter the empty space in the joints were filled with the suitable nanofillers doped adhesive followed by drying for 24 hours at room temperature and then in 6 hours at 60 °C in the oven. Each joint type made consisted of 3 samples as shown in Figure 4.

2.6. Procedure for Tensile test

A computer controlled Universal Testing Machine (UTM) operating at room temperature was used for conducting the tensile test. The joint sample dimensions were maintained according to ASTM D-5868 standards as depicted in Figure 2. The joint samples were categorized into 10 groups with 3 SLJ specimens in each group as given in Table 1; the SLJ specimens were loaded into the UTM and were
tested at a feed rate of 1 mm/min as shown in Figure 5. Upon successful completion of the test, the joints were perceived for cohesive and adhesive damage.

![Figure 5. Tensile test set up for different joint composition.](image)

### Table 1. Labeling and composition of Joints samples.

| Joint Designation | Adherent/Adhesive/Reinforcement |
|-------------------|---------------------------------|
| C/AD 0.5/I        | C/ AD -0.5mm/Non reinforced     |
| C/AD -1/I         | C/AD - 1mm/Non reinforced       |
| C/AD -0.5/CNT/II  | C/ AD -0.5mm/MWCNT/1%           |
| C/AD -0.5/F/II    | C/ AD -0.5mm/Fullerene/1%       |
| C/AD -1/CNT/III   | C/ AD -1mm/MWCNT/1%             |
| C/AD -1/F/III     | C/ AD -1mm/Fullerene/1%         |
| C/AD -0.5/CNT/IV  | C/ AD -0.5mm/MWCNT/3%           |
| C/AD -0.5/F/IV    | C/ AD -0.5mm/Fullerene/3%       |
| C/AD -1/CNT/V     | C/ AD -1mm/MWCNT/3%             |
| C/AD -1/F/V       | C/ AD -1mm/Fullerene/3%         |

### 3. Results & Discussion

Average damage load as given in Table 2 was obtained for each joint composition as mentioned in Table 1 by conducting the tensile test. Each category had 3 samples. It was noted that the average failure load for the non doped joint configuration was obtained to be least among all when compared with 1 wt % and 3 wt % nanofillers doped joints. This is because the addition of nanofillers increases the capacity for displacement which makes the nanofillers doped joints capable of taking more loads before failure. Also a decrease of 12.51% was observed in the damage load as the thickness of the adhesive layer increased from 0.5 mm to 1 mm. For an adhesive thickness of 0.5 mm, an increase of 82% and 154% in the joint strength was observed when MWCNT was added at 1 wt % and 3 wt %. Similarly for 0.5 mm adhesive thickness, the joint strength increased by 49% and 33% on addition of 1
wt % and 3 wt % of Fullerene C$_{60}$ respectively. This can be seen from the variation of average failure load in Figure 6.

Table 2. Average failure load for each composition of joints.

| Joint Designation | Average Failure Load (N) |
|-------------------|--------------------------|
| C/AD 0.5/I        | 1782                     |
| C/AD-1/I          | 1559                     |
| C/AD-0.5/CNT/II   | 3254                     |
| C/AD-0.5/F/II     | 2656                     |
| C/AD-1/CNT/III    | 2736                     |
| C/AD-1/F/III      | 2259                     |
| C/AD-0.5/CNT/IV   | 4525                     |
| C/AD-0.5/F/IV     | 2364                     |
| C/AD-1/CNT/V      | 3784                     |
| C/AD-1/F/V        | 1883                     |

Figure 6. Average failure loads for various joint compositions at 0.5 mm adhesive thickness.

Similarly when the adhesive thickness was increased to 1mm keeping the weight percentage of MWCNT and Fullerene C$_{60}$ at 1 wt % and 3 wt %, a rise of 75% and 143% for MWCNT doped joints while 45% and 21% for Fullerene C$_{60}$ doped joints was procured. These are indicated from the average failure load given in Figure 7.
Figure 7. Average failure loads for various joint compositions at 1 mm adhesive thickness.

The highest average damage load for MWCNT was attained at 3 wt % with 0.5 mm adhesive thickness. But for the same composition at 1 mm adhesive thickness, the average damage load decreased by 16%. Likewise for Fullerene C$_{60}$ the highest average damage load was attained at 1 wt % with 0.5 mm adhesive thickness while with the increment in the adhesive thickness to 1 mm, the damage load decreased by 15%. The average damage load obtained for all category of specimen was high when the adhesive thickness was less. This was because the SLJ specimens had ability to carry higher load due to the ductile nature of the joints. But with the increase in the adhesive thickness the average damage load for non-doped, MWCNT and Fullerene C$_{60}$ doped joints decreased. This was due to the brittleness induced at the place of joint formation. A dip in the average damage loading was observed for both the 1 wt % and 3 wt % and Fullerene C$_{60}$ doped joints when the thickness of the adhesive layer was increased. This is because the increase in adhesive layer thickness makes the joints brittle which is not capable of carrying greater load.

4. Conclusion
This work was focused on variation in adhesive layer thickness and variation in weight percentage of nanofillers on the SLJ’s under tensile loading. The conclusion drawn out is as follows:

- The failure of the joints and their load carrying capability depends on the thickness of the adhesive layer. With the increase in thickness of the adhesive layer, the joints became brittle in nature thereby reducing the joint strength.
- The addition of nanofillers and its dispersion into the adhesive raised the load carrying ability of the joints when compared with non doped joints.
- Addition of MWCNT in 1 wt % and 3 wt % leads to improvement in the load bearing capacity of the joints. The highest load bearing ability of the joint is seen at 3 wt %. But when the thickness of the adhesive layer was increased from 0.5mm to 1mm the load bearing ability took a dip.
- Addition of Fullerene C$_{60}$ in the adhesive indicated greater average damage load carrying capacity at 1 wt % but when the wt % of Fullerene C$_{60}$ was raised to 3, a decrease in the damage load carrying capacity was observed.
5. Reference

[1] M D Banea and L F M da Silva 2008 Adhesively bonded joints in composite materials: an overview Journal of Materials: Design and Applications, Proc. IMechE 223

[2] Ferhat K and Hasan P 2016 Effects of Different Fiber Orientations on the Shear Strength Performance of Composite Adhesive Joints International Journal of Chemical, Molecular, Nuclear, Materials and Metallurgical Engineering 10 65-68

[3] F Kadioglu, E Avil, M E Ercan, T Aydogan 2018 Effects of different overlap lengths and composite adherend thicknesses on the performance of adhesively-bonded joints under tensile and bending loadings, IOP Conf. Series: Materials Science and Engineering 369 012034

[4] Ashby M F, Ferreira P J and Schodek D 2009 Nanomaterials, nanotechnologies and design: an introduction for engineers and architects Amsterdam: Elsevier/Butterworth Heinemann

[5] B Soltannia and F Taheri 2015 Influence of nano-reinforcement on the mechanical behavior of adhesively bonded single-lap joints subjected to static, quasi-static, and impact loading Journal of Adhesion Science and Technology 29 424-442

[6] Kanu Priya Jhanji, Asokan R, Sharanyaa Pandima Dhevi S and Mahathi Guptha K 2019 Effect of Healing Agent Filled Microcapsules on Tensile Strength of Glass/ Epoxy Composite Laminate International Journal of Vehicle Structures & Systems 11 250-254

[7] A J Kinloch, J H Lee, A C Taylor, S Sprenger, C Eger and D Egan 2003 Toughening structural adhesives via nano- and micro-phase inclusions Journal of Adhesion 79 867-873

[8] Almir Silva Netoa, Diego Thadeu Lopes da Cruz and Antonio Ferreira Ávilab 2013 Nano-modified Adhesive by Graphene: The Single Lap-Joint Case Materials Research 16 592-596

[9] Liberata Guadagno, Maria Sarno, Umberto Vietri, Marialuigia Raimondo, Claudia Cirillo and Paolo Ciambelli 2015 Graphene-based structural adhesive to enhance adhesion performance, RSC Advances 5 27874-27886

[10] Michael B J, B Ashrafi, Yunfa Zhang, Yadienka M R, Christopher T K, Andrew Johnston and B Simard 2015 Single-walled carbon nanotube–epoxy composites for structural and conductive aerospace adhesives Composites: Part B 69 87–93

[11] V K Srivastava 2011 Effect of carbon nanotubes on the strength of adhesive lap joints of C/C and C/C–SiC ceramic fibre composites International Journal of Adhesion & Adhesives 31 486-489

[12] Kuang-Ting Hsiao, Justin Alms and Suresh G A, 2003 Use of epoxy/multiwalled carbon nanotubes as adhesives to join graphite fibre reinforced polymer composites Nanotechnology 14 791–793

[13] R Amit Kumar, R Asokan, Kanu Priya Jhanji, Debajit Das and N Vishnu Sai 2019 Investigation of Tensile Properties of Carbon/Epoxy Composite Joints with and without Carbon Nanotubes International Journal of Vehicle Structures & Systems 11 209-213

[14] V E Muradyan, A A Arbuzov and Yu N Smirnov 2009 Adhesive Strength of Fullerene-Doped Epoxyamine Compositions Russian Journal of General Chemistry 79 797–799