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Influence of adhesive thickness and filler content on the mechanical performance of aluminum single-lap joints bonded with aluminum powder filled epoxy adhesive

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ABSTRACT

The objective of this study was to develop information on the influence of adhesive thickness and aluminum filler content on the mechanical performance of aluminum joints bonded by aluminum powder filled epoxy. The adhesive strength of the joints was determined by utilizing the single-lap shear test. The influence of adhesive thickness and aluminum filler content on stress distribution within the adhesive was also analyzed by finite element method (FEM). Both FEM analysis and the experimental investigation show that in general adhesion strength decreases as the thickness of the adhesive increases. It is observed from the predictions (FEM simulations) that the stress level increases at the adhesive–metal substrate interface as the aluminum filler content in the adhesive increases. Experimental results show that epoxy adhesive retains its strength up to the 50 wt% aluminum filler content. The joints fail in cohesive mode (failure within the adhesive) due to the high stress levels generated in the adhesive, which indicates that the adhesion to the metal surface is stronger than that of the interior part of the adhesive.

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1. Introduction

Epoxy resins are widely used as high-performance structural adhesives, especially in automotive and aircraft manufacture. Epoxy resins are attractive for metal-bonding adhesive systems because of their ability to cure without producing volatile by-products and their low shrinkage upon curing (less than 0.5%) (Tai and Szklarska-Smialowska, 1993). Epoxy resins are able to bond well to a variety of treated or untreated metal surfaces (Mohan, 1990). In aircraft manufacture, there is a great need for evenly stressed, smooth bonding of thin aluminum sheet and honeycomb materials. Epoxy adhesives have a good affinity for aluminum alloy surfaces, and the oxide layers produced during surface preparation (Chasser et al., 1993).

To be able to obtain a strong and stable bond between the metal and the adhesive, the natural surface oxide should be removed and replaced with a new, continuous, solid, corrosion resistant oxide layer. The removal can be done mechanically and/or chemically. Mechanical (abrasive) cleaning also increases the surface roughness and, consequently, the bond strength by mechanical interlocking and by the increased number of chemical bonds on the larger surface area. Various chemical treatments, the most common being acid etches, have been developed to modify the oxide, to render it more receptive to bonding (Kozma and Olefjord, 1987a,b; Brewis, 1985; Hogg and Janardhana, 1993; Kinloch, 1983).

However, often in industrial practice, although the benefits of surface preparation are widely known, it is not done

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properly for economic and safety reasons. Proper surface preparation takes extra production time, tends to be labor intensive and usually involves use of hazardous materials. Hence, an industrial structural adhesive must be suitable for use on less than ideal surfaces.

In a variety of industrial applications epoxy adhesives are required to have an enhanced thermal conductivity. The normal method for changing this physical property is to add to the epoxy a filler of higher conductivity than the continuous phase (Tai and Szklarska-Smialowska, 1993; Hermansen and Tunick, 1989; Tomlinson and Stapley, 1977; Lee and Neville, 1967; Kingery, 1960; Nieberlein and Steverding, 1977; Gaynes et al., 1997; Hahn et al., 1998; Subramanian et al., 1998; Nikkeshi et al., 1998). By the incorporation of fillers into the adhesive, the resin content (and thus the cost) is also reduced.

Achieving improved thermal conductivity is dependent on filler selection and loading level. Filler type, size, shape and volume fraction determine the adhesive thermal conductance. The adhesive thermal conductivity increases with increasing volume fraction of the filler. Theoretically, the thermal conductivity of the filler is not an important variable except when it is within a factor of 10 of the thermal conductivity of the polymeric matrix (adhesive). Most metal fillers have thermal conductivities greater than 10 times the matrix thermal conductivity (Hermansen and Tunick, 1989).

Alumina powder is one of the commonly used fillers for improving the thermal conductivity of adhesives, in particular insulation adhesives. Aluminum and silver powders or flakes are used to improve the thermal and electrical conductivities for adhesives intended to be an electrical or thermal path (Hermansen and Tunick, 1989; Kang and Purushothaman, 1998; Lu et al., 1999). The filler level must be sufficiently high to achieve point-to-point contact before electrical conductivity is attained (Hermansen and Tunick, 1989). However, too high filler content might cause a degradation in mechanical properties of the adhesive (Nikkeshi et al., 1998). There are also several commercially available epoxy adhesives reinforced with other metal fillers such as aluminum powder.

While the improvement on thermal properties of adhesives by addition of metal fillers is obvious (Hermansen and Tunick, 1989; Tomlinson and Stapley, 1977; Lee and Neville, 1967; Kingery, 1960; Nieberlein and Steverding, 1977), their influence on the mechanical properties of the adhesive joints are not clear. The results of this study are expected to shed light on this aspect. The objective was to investigate the influence of the filler content and the adhesive thickness on the mechanical performance of aluminum single-lap joints bonded with aluminum powder filled epoxy adhesive.

The traditional evaluation of adhesive joints by strength measurements was utilized in the study. The adhesive strength was determined by utilizing the single-lap shear test (Mohan, 1990; Arnold, 1989; Tsai and Morton, 1994; Annual Book of ASTM Standards, 1992). The single-lap joint configuration (Fig. 1) is widely used in the aerospace, automotive and wood and plastic industries (Hermansen and Tunick, 1989; Arnold, 1989; Stringer, 1985; De Wilde et al., 1995; Jialanella and Shaffer, 1993). Single-lap specimens are economical, practical and easy to make. They also allow easy control (and measurement) of the bond thickness.

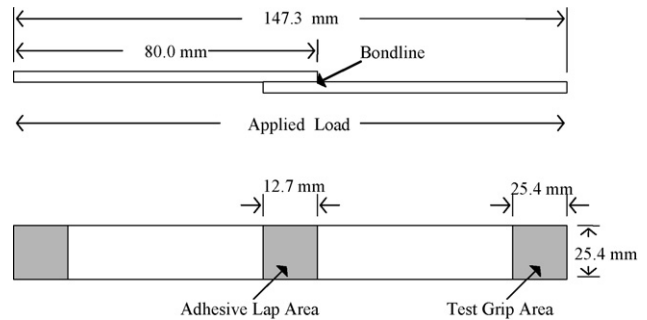


Fig. 1 – Single-lap shear joint configuration.

The variation of adhesion strength with adhesive thickness is examined in this paper. The effect of bond thickness on adhesive performance for various other joints has been reported by few investigators (Stringer, 1985; Foulkes et al., 1970; Hylands, 1984; Beevers, 1986). However, there is a lack of study on the variation of adhesion characteristics with adhesive thickness for single-lap shear joints. Consequently, in the present study, the effects of metal filler addition and the adhesive thickness on stress distribution within the adhesive joints are analyzed by the finite element method. The experiments are also carried out to validate and compare the theoretical predictions of the adhesion strength of adhesive bonds at different thicknesses.

2. Experimental

2.1. Materials

The epoxy adhesive used in this investigation is a general-purpose, two-part epoxy (Fusor 309) obtained from Lord Corporation. The adhesive is prepared by mixing equal volumes of the resin and hardener parts. The mixed adhesive cures fully in 24–48 h at room temperature with handling strength in about 8 h.

The aluminum powder used for filling the epoxy adhesive was obtained from Allied Britannia Limited. The aluminum particles (filler) were spherical/roundish with size smaller than 50 μm in diameter.

The aluminum sheets used as adherents in making aluminum joints were cut from locally obtained aluminum plates.

2.2. Adhesive joint preparation

The following procedures were used for cleaning the aluminum sheets (80 mm \times 25.4 mm) before adhesively joining them (Kozma and Olefjord, 1987a; Prakash et al., 1987; Semerdjiev, 1970).

1. Degrease by dipping shortly in trichloroethylene and isopropyl alcohol, separately.
2. Wash with water.
3. Roughen surfaces by abrader cleaning (mechanical cleaning) by 400 grit silicon carbide grinding paper.
4. Degrease by dipping in trichloroethylene and isopropyl alcohol (30 min each).

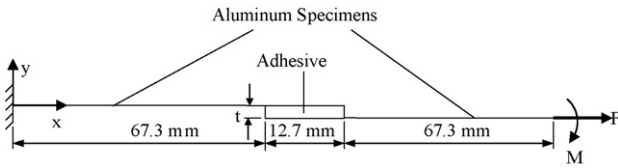


Fig. 2 – Finite element model of the system.

5. Immerse for 2–4 h in a solution of H₂SO₄, sodium dichromate and distilled water in proportion: 22.5, 7.5 and 70 by weight, respectively (chromic–sulphuric etching process).
6. Wash with distilled water.
7. Dry with clean paper or tissue and keep in a desiccator until use.

The single-lap shear adhesive joints were then prepared by bonding surface cleaned/treated aluminum sheets together with neat epoxy adhesive and then after addition of aluminum powder at several fractions (10, 25 and 50 wt%). Equal volumes of the epoxy resin and the hardener were mixed and then aluminum powder was added into the adhesive mixing again. A designed fixture was used to assemble the adhesive joints. It had two fixed end plates and a movable one between them. The metal sheets were bonded together between the movable plate and the far end plate. The fixture included a micrometer used for controlling the adhesive thickness. The actual adhesive thickness of the cured joint was measured by a Digimatic Caliper.

2.3. Mechanical characterization of the adhesive joints

The adhesive joints were assembled and tested as specified in ASTM D 1002 (Annual Book of ASTM Standards, 1992). The joint configuration is shown in Fig. 1 while the applied loads are shown in Fig. 2. End tabs of the same thickness and material of the adherends were used to place the load axis in the same plane of the overlap area. The equipment used for mechanical characterization was an Instron 5567 mechanical testing system. Joint strengths, reported in units of shear stress, were calculated as follows:

$$\text{joint strength} = \frac{\text{failure load}}{\text{adhesive lap area}} \quad (1)$$

3. Stress analysis by finite element method

The influence of metal filler content and the adhesive thickness on stress distribution within the adhesive joints was analyzed by finite element method (FEM). Due its versatility and computational power, the FEM is commonly used in the analysis of modern engineering and scientific systems. In this study, the FEM is used to model and analyze the adhesive joint through the well-known finite element package ANSYS. The system consisting of the joint and two-bonded aluminum specimens is subjected to tension. The resulting shear and von Mises stresses are computed for various adhesive compositions and thicknesses to see their effects on the mechanical strength of the joint. These stresses are chosen, because it is

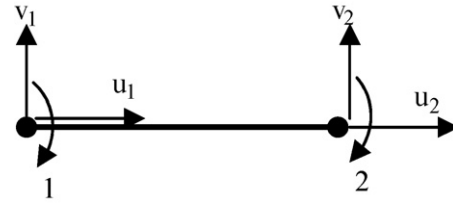


Fig. 3 – The elastic beam element.

expected that the shear stress is the mode of failure for the adhesive joint and the von Mises stress is a measure on the state of equivalent stress.

The finite element model of the system is shown in Fig. 2. The two aluminum specimens seen in the figure are modeled using the BEAM3 element in ANSYS (ANSYS, 2005). Depicted in Fig. 3, this elastic beam element allows in-plane horizontal and vertical displacements (u and v) and rotation about the out-of-plane axis (θ) at the two end nodes of 1 and 2. As shown in Fig. 3, the gripped portion of the left specimen is fixed and the right specimen is pulled with a force (F) of 100 N resembling the actual testing conditions. In the actual test, this force is aligned with the central axis of the joint and hence a bending moment of $M = Ft/2$ is applied to account for the eccentricity of the force at the finite element model (where t is the adhesive thickness).

The two dimensional (2D) quadrilateral element with mid-side nodes and thickness option, PLANE82, was used for modeling the adhesive bond giving in-plane displacements at the eight nodes. This element is a higher version of the four-noded element in ANSYS and is better suited for the applications where the bending moment exists (ANSYS, 2005). The adhesive bond is made of epoxy and aluminum filler whose weight percentages are changed together with the bond thickness (t) to observe their effects on the mechanical strength of the joint. The material properties for the aluminum and epoxy are listed in Table 1. The modulus of elasticity (E) and Poisson's ratio (ν) for the adhesive are assumed as

$$E = c_a E_a + c_e E_e \quad (2)$$

$$\nu = c_a \nu_a + c_e \nu_e \quad (3)$$

where E_a and E_e are the moduli of elasticity, and ν_a and ν_e are the Poisson's ratios for the aluminum and epoxy, respectively. The coefficients c_a and c_e in the above equations denote the fractions corresponding to the aluminum filler and epoxy.

Table 1 – Material properties

	Aluminum	Epoxy
Modulus of elasticity (E, GPa)	68.95	1.0858
Poisson's ratio (ν)	0.333	0.38

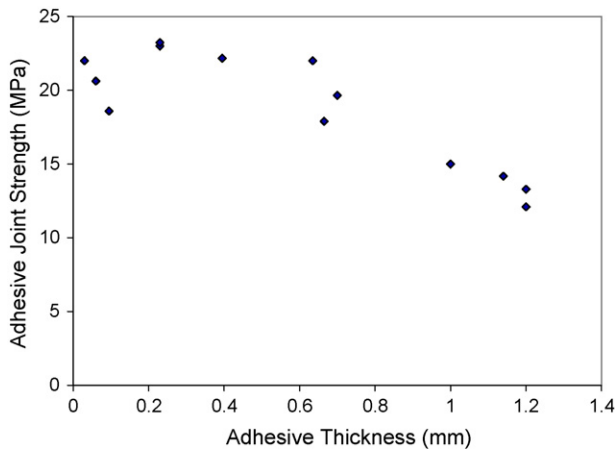


Fig. 4 – A plot of adhesive joint strength vs. adhesive thickness for aluminum joints bonded with neat epoxy with no filler.

4. Results and discussion

4.1. Mechanical characterization

Adhesive joint strength versus adhesive thickness is plotted in Fig. 4 for aluminum joints bonded with neat epoxy with no filler content. Adhesive thicknesses investigated ranged from 0.03 mm to 1.3 mm. As seen in the plot there is a general trend of decrease in single-lap adhesive joint shear strength with adhesive thickness. Increase of adhesive thickness from 0.03 mm to 1.3 mm resulted a decrease of about 35–40% in adhesive joint shear strength. However the effect was not significant for adhesive thicknesses up to about 0.7 mm.

In light of the above result, the smallest adhesive thickness (0.03–0.05 mm) was used for the experiments of the rest of the investigation.

A plot of adhesive joint strength versus amount of aluminum powder used in the adhesive is presented in Fig. 5. Addition of aluminum particles as much as 50 wt% did not cause a significant decrease in adhesive joint strength. It

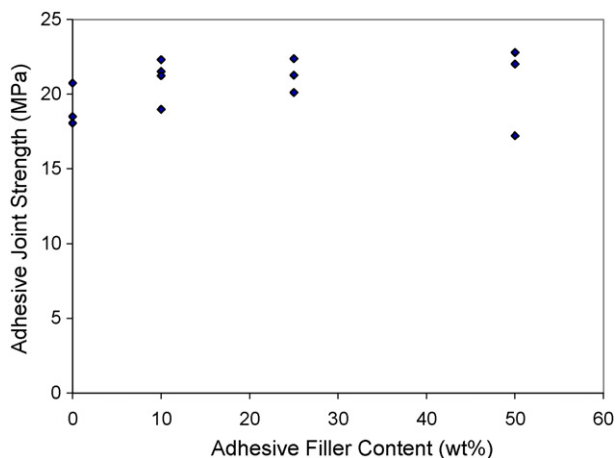


Fig. 5 – A plot of adhesive joint strength vs. adhesive filler content.

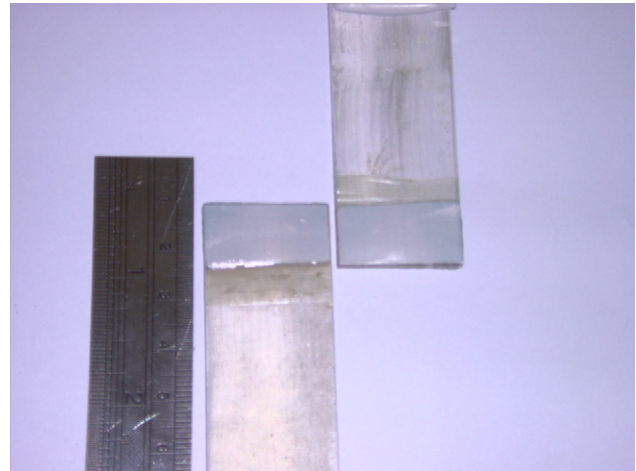


Fig. 6 – Fracture surfaces of an aluminum joint bonded by epoxy with no metal filler.

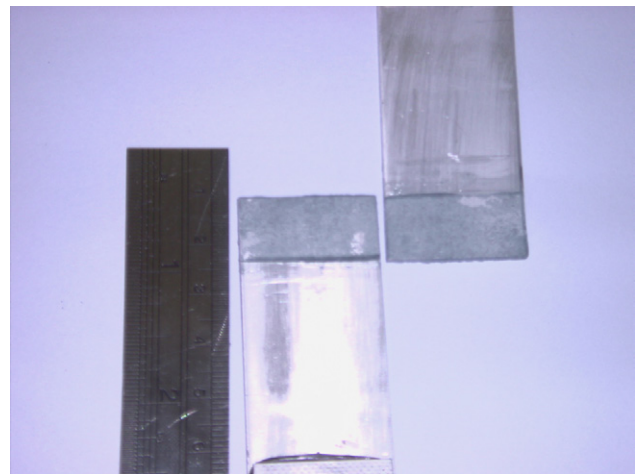


Fig. 7 – Fracture surfaces of an aluminum joint bonded by epoxy with 25 wt% aluminum filler.

should be noted that 50 wt% Al filler content in the adhesive corresponds to about 44.45% in volume because of density difference between aluminum particles and epoxy adhesive.

Fractured specimens were visually examined to determine the failure mode. The joints failed almost completely in cohesive failure mode. Cohesive failure occurs when the adhesive separates from itself (failure within adhesive). Adhesive layers left on both joint surfaces are clearly shown in Figs. 6 and 7 for neat epoxy and epoxy with 25 wt% aluminum filler content, as examples.

4.2. FEM analysis

The shear and von Mises stresses for various bond thicknesses and various adhesive compositions were analyzed. According to the stress contours which are not shown here, the maximum shear and von Mises stresses occur at the adhesive–metal substrate interface. This is especially true for the shear stresses, which almost vanish toward the middle of the adhesive. The same trend also exists for the von Mises

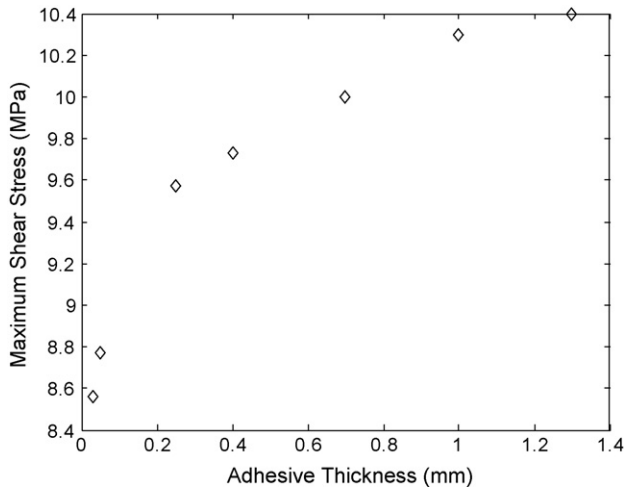


Fig. 8 – Maximum shear stress vs. adhesive thickness for neat epoxy.

stresses, which attain maximum at the edges and decrease away from the edges.

In order to investigate the effect of bond thickness on the bond strength, the maximum shear and von Mises stresses are plotted against the bond thickness for neat epoxy adhesive in Figs. 8 and 9. As seen in these figures shear stresses in the adhesive increase (decreasing the bond strength) as the bond thickness increases. As for the maximum von Mises stresses, they show slightly different or mixed behavior (Fig. 9), but the general trend is similar to the trend for the shear stresses. In other words, the von Mises stresses are also increased as the thickness of the bond is increased. It should be noted that the shear stress contribution to the von Mises stress is significant in the bond region close to the aluminum plate. This, in turn, results in possible failure of bonding in this region.

Fig. 10 shows the plots of finite element predictions and experimental results for the adhesive joint strength of neat epoxy at different adhesive thicknesses. It can be

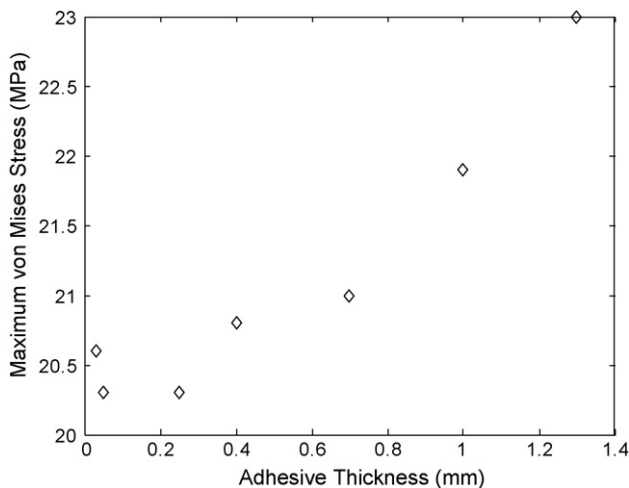


Fig. 9 – Maximum von Mises stress vs. adhesive thickness for neat epoxy.

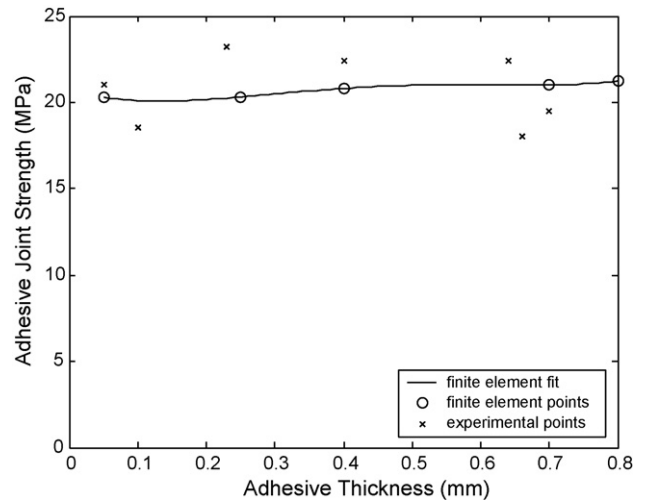


Fig. 10 – Adhesive joint strength vs. adhesive thickness for neat epoxy.

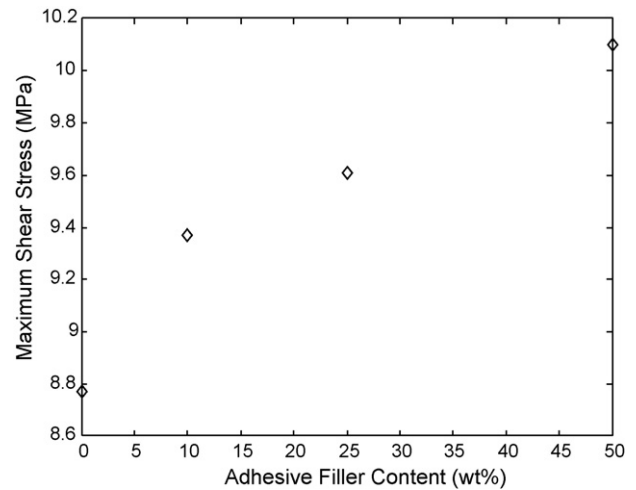


Fig. 11 – Variation of maximum shear stress with aluminum content for adhesive thickness of 0.05 mm.

observed that the finite element predictions agree well with the experimental results. Some discrepancies between both results are because of (i) the experimental error, which is estimated as 7% (based on the experimental repeats) and (ii) the assumption of the homogeneous adhesive properties across the bonding section in the finite element simulations.

Stress analysis was also done for various adhesive compositions. The mass average of mechanical properties was used in simulations to account for the aluminum powder concentration in the adhesive composition. Figs. 11 and 12 show the plots of the maximum shear and von Mises stresses versus adhesive compositions for the bond thickness of 0.05 mm. It is evident that these stresses get higher as the weight percentage of aluminum content in the adhesive bond increases. In other words, the aluminum content in the adhesive adversely affects the adhesion strength of the joint. However, as the bond thickness increases this effect diminishes and the rate of increase in stresses tends to decrease at high bond thick-

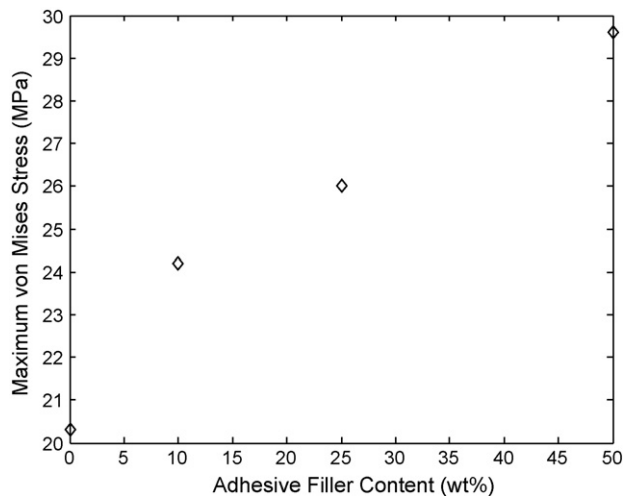


Fig. 12 – Maximum von Mises stress vs. aluminum content for adhesive thickness of 0.05 mm.

nesses. Results for the adhesive thickness of 1.0 mm are presented in Figs. 13 and 14.

Furthermore, as stated in the manuscript in Section 3, the shear stress is the possible cause of failure for the adhesive joint. Hence, as seen in Figs. 11 and 13, the maximum shear stress is, in general, higher for the adhesive thickness of 1 mm than that corresponding to the thickness of 0.05 mm. This result indicates the adverse effect of the adhesive thickness increase in bond strength.

Although in the present study, aluminum powder is used as filler in the adhesive, investigation into mechanical response of the joint when other powder such as copper will be fruitful for future investigation. In addition, examination of the mechanical response of the joints due to mixture of aluminum and copper powders used as filler in the adhesive will be interesting for improved thermal and electrical conductivities.

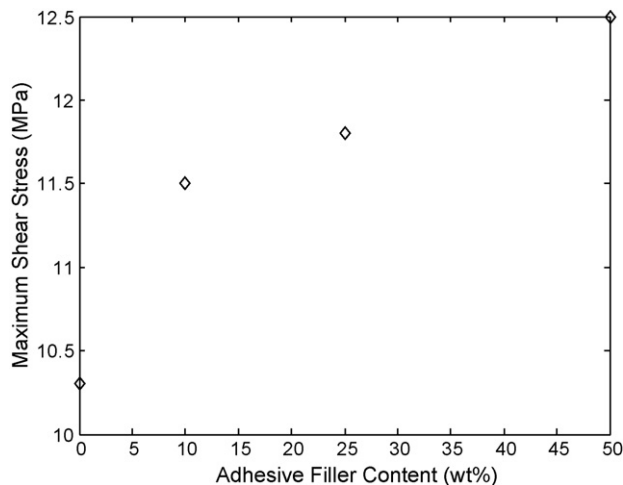


Fig. 13 – Variation of maximum shear stress with aluminum content for adhesive thickness of 1 mm.

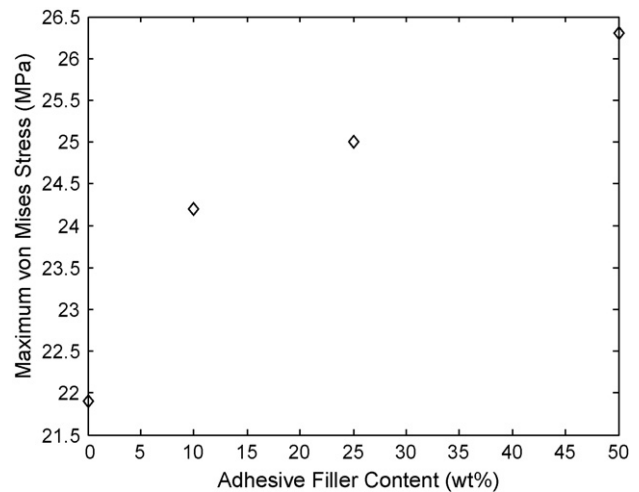


Fig. 14 – Maximum von Mises stress vs. aluminum content for adhesive thickness of 1 mm.

5. Conclusions

The following statements can be made from the findings of this study:

1. According to the results of both FEM analysis and the experimental investigation, adhesive thickness has a negative effect on single-lap adhesive joint shear strength.
2. The finite element results on stresses show that the aluminum content in the adhesive adversely affects the mechanical strength of the bond.
3. However, a promising result was obtained through experimental investigation that epoxy adhesive retains its adhesion strength even with as much as 50 wt% addition of aluminum filler.
4. Even though the finite element analysis shows higher stresses at the adhesive–metal substrate interface, actual failure occurs within the adhesive indicating that the strength of adhesion to the metal substrate surface is stronger than the strength of the adhesive itself.

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REFERENCES

- Annual Book of ASTM Standards, 1992. American Society for Testing and Materials, vol.15.06. ASTM, Philadelphia, PA, pp. 47–50.
- ANSYS, 2005. Release 10.0, ANSYS, Inc., Canonsburg, Pennsylvania, USA.
- Arnold, J.R., July 1989. Adhesively bonded coated steels. *Automot. Eng.*, 39–44.
- Beevers, A., 1986. Coaxial joints in engineering assembly. *Mater. Sci. Technol.* 2, 97–102.

- Brewis, D.M., 1985. Factors affecting bonding of metals. In: Continuous Casting '85, Proceedings of the International Conference Sponsored and Organized by The Institute of Metals, London, pp. 629–644.
- Chasser, A.M., Makhlof, J.M., Schneider, J.R., March 1993. Rubber-based structural adhesive is a new option for metal bonding. *Adhes. Age*, 36–39.
- De Wilde, W.P., VanVinckenroy, G., Tirry, L., Cardon, A.H., 1995. Effects of the environment and curing on the strength of adhesive joints. *J. Adhes. Sci. Technol.* 9, 149–158.
- Foulkes, H., Shields, J., Wake, W.C., 1970. The variation of bond strength with temperature: a preliminary study of metal-to-metal adhesion. *J. Adhes.* 2, 254–269.
- Gaynes, M.A., Matienzo, L.J., Zimmerman, J.A., Vanchart, D., 1997. Analysis and characterization of electrically conductive adhesives. In: *Electronic Packaging Materials Science. IX. Materials Research Society Symposium Proceedings*, Warrendale, PA, USA, pp. 139–151.
- Hahn, O., Meschut, G., Koyro, M., 1998. Thermal conductivity, strength and ageing behaviour of adhesive-bonded joints with filler-modified adhesives. *Schweissen und Schneiden/Weld. Cutting* 50 (7), E130–E133.
- Hermansen, R.D., Tunick, S.A., October 1989. Formulating custom-tailored thermal transfer adhesives. *Adhes. Age*, 38–41.
- Hogg, I.C., Janardhana, M.N., 1993. Surface preparation of adhesively bonded joints. In: Chandra, T., Dhingra, A.K. (Eds.), *Advanced Composites '93, International Conference on Advanced Composite Materials. The Minerals, Metals & Materials Society*, pp. 455–461.
- Hylands, R.W., 1984. Strength characteristics of mono and multiple-wire steel to steel joints bonded with an epoxy adhesive. In: Mittal, K.L. (Ed.), *Adhesive Joints: Formation, Characteristics, and Testing*. Plenum Press, New York, pp. 165–193.
- Jialanella, G.L., Shaffer II, E.O., 1993. The effect of adhesive modulus on the performance of SMC lap shear joints. *J. Adhes. Sci. Technol.* 7, 1171–1181.
- Kang, S., Purushothaman, S., 1998. Development of low cost, low temperature conductive adhesives. In: *Proceedings of the Electronic Components and Technology Conference, IEEE, Piscataway, NJ, USA*, pp. 1031–1035.
- Kingery, W.D., 1960. *Introduction to Ceramics*. John Wiley & Sons, Inc., New York.
- Kinloch, A.J., 1983. *Durability of Structural Adhesives*. Applied Science Publishers, London.
- Kozma, L., Olefjord, I., 1987a. Basic processes of surface preparation and bond formation of adhesively joined aluminum. *Mater. Sci. Technol.* 3, 860–874.
- Kozma, L., Olefjord, I., 1987b. Surface treatment of steel for structural adhesive bonding. *Mater. Sci. Technol.* 3, 954–962.
- Lee, H., Neville, K., 1967. *Handbook of Epoxy Resins*. McGraw-Hill, New York.
- Lu, D., Tong, Q.K., Wong, C.P., 1999. Conductivity mechanisms of isotropic conductive adhesives (ICA's). *IEEE Trans. Electron. Pack. Manuf.* 22 (3), 223–227.
- Mohan, R., February 1990. Analyzing adhesively bonded joints for automotive applications. *Plast. Eng.*, 47–51.
- Nieberlein, V.A., Steverding, B., 1977. Thermal conductivity of epoxy–aluminum powder mixtures. *J. Mater. Sci. Lett.* 12, 1685–1688.
- Nikkeshi, S., Kudo, M., Masuko, T., 1998. Dynamic viscoelastic properties and thermal properties of Ni powder–epoxy resin composites. *J. Appl. Polym. Sci.* 69 (13), 2593–2598.
- Prakash, R., Srivastava, V.K., Gupta, G.S.R., December 1987. Behavior of adhesive joints in corrosive environment. *Exp. Mech.*, 346–351.
- Semerdjiev, S., 1970. *Metal-to-Metal Adhesive Bonding*. Business Books Limited, London.
- Stringer, L.G., 1985. Comparison of the shear stress–strain behaviour of some structural adhesives. *J. Adhes.* 18, 185–196.
- Subramanian, S., Kustas, F., Rawal, S., Shinn, E.T., 1998. Fabrication, testing and analysis of carbon-carbon/aluminum bonded joints with high thermal conductivity. In: *Collection of Technical Papers—AIAA/ASME/ASCE/AHS/ASC Structures, Structural Dynamics & Materials Conference*, vol.3, AIAA, Reston, VA, USA, pp. 2423–2433.
- Tai, R.C.L., Szklarska-Smialowska, Z., 1993. Absorption of water by different fillers-incorporated automotive epoxy adhesives. *J. Mater. Sci.* 28, 6199–6204.
- Tomlinson, W.J., Stapley, D., 1977. Thermal conductivity of epoxy resin–aluminum (0–50%) composites. *J. Mater. Sci. Lett.* 12, 1689–1690.
- Tsai, M.-Y., Morton, J., 1994. A note on peel stresses in single-lap adhesive joints. *J. Appl. Mech.* 61, 712–715.