

The Effect of Polymer Network Structure Upon the Bond Strength of Epoxy-Aluminum Joints

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Synopsis

The bond strength to aluminum of Epon 828 cured with various amounts of methylene dianiline has been investigated as a function of the resin network structure. In order to meet this objective, a torsional test for bond shear strength was developed, and fully cured resins with different network structures were prepared. The effects of the rate of loading of the joint and the aluminum surface pretreatment on the bond strength were also examined. Very high bond shear strengths, in excess of 9000 psi, were found for joints which after machining had been polished, vapor degreased, and treated with dilute sulfuric acid-potassium dichromate solution. It was found for these joints that the average bond strength decreased as the molecular weight between cross-links increased. For the joints without acid-dichromate treatment, the failures were adhesive, and the network structure did not seem to significantly affect the bond strength. There are indications that chemical bonding occurred in the case of the acid-dichromate-treated joints; the decrease in bond strength as per cent excess amine and M_c increased is possibly associated with a decreased amount of chemical bonding. The bond strength increased to a limiting value as the rate of testing increased.

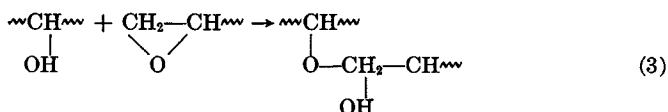
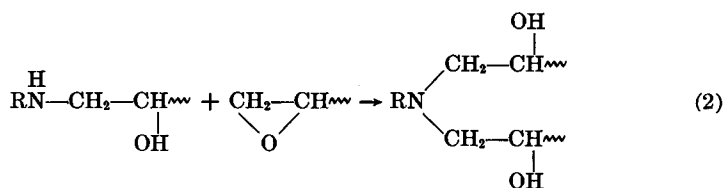
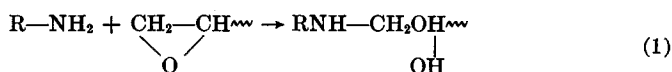
INTRODUCTION

Epoxy resin adhesives can be used to bond parts of geometry such that they would be difficult to fasten together otherwise, and the epoxy resin imposes very little additional weight. In the aerospace industry, where aircraft of ever-increasing size and geometric complexity are a way of life, epoxy resin bonding of nonload-bearing or low load-bearing parts is a standard procedure. One would like also to be able to use epoxy bonding systems for direct load-bearing and structural applications, but this has been avoided to a considerable extent because of high variability of the bond strengths. This is in turn due to a general lack of knowledge about both the structure of the cured resin and the factors that affect the epoxy resin-metal bond. Applications have been developed on a purely empirical basis, with the criteria for success or failure usually being rather arbitrary. Since several polymer structure variables which are thought to be important are uncontrolled or poorly controlled, the variability in the final product is not surprising.

The bond preparation procedure which is generally used involves mixing the epoxy compounds with a reactant which makes polymerization and

crosslinking possible. Physical properties vary with the degree of crosslinking. In order to polymerize and then crosslink, the mixture of epoxy resin and hardener is heated for a given time at a given oven temperature, and finally heated for another period of time at a higher oven temperature. Since the polymerization-crosslinking reaction is highly exothermic, and since there is a definite induction period which can be related to small and variable impurity concentrations, control of the time and oven temperature is insufficient to control the course of the reaction. The relative and absolute amounts of crosslinking and polymerization in turn control the network structure and the concentration of functional groups available to interact with the metal. Control of the process is at present difficult, but it is possible in at least some cases to determine the course and extent of the reaction at any given time by the use of the analysis for epoxy and amine groups and mathematical techniques which have been recently developed.¹

Schechter et al.² have investigated epoxy group reactions with amines. They suggested three possible reactions as shown below:



Schechter et al.² found that the amine reactions (1) and (2) predominate, and the amount of reaction (3) occurring could hardly be distinguished from experimental error. Reaction (3) would be expected to be accelerated by the presence of a strong tertiary amine, but Schechter et al. attribute the absence of a major reaction catalyzed by the tertiary amine formed in reaction (2) to the steric hindrance of this amine. The curing reaction can therefore often be visualized as occurring in two steps: reaction (1) followed by reaction (2). Kinetic data show that reaction (1) is about ten times more rapid than reaction (2).

Consider a moles of diamine to be mixed with b moles diepoxide, and suppose that at a given time there are X unreacted primary amine RNH_2 groups and Y secondary R_2NH groups. This means that $2a - X$ primary H and $2a - X - Y$ secondary H atoms have reacted, so that the concentration of unreacted epoxy groups, E is

$$E = 2b - (2a - X) - (2a - X - Y) = 2b - 4a + 2X + Y.$$

If the experiment is carried out so that X is measured and either no epoxy groups remain or the epoxy content is measured, one may calculate Y . The concentration of secondary amine which has reacted is then $2a - X - Y$, which is also the number of branch points. The total number of chain segments will be $^{3/2}(2a - X - Y) + [(X + E)/2]$.

The equation for the effective M_c is

$$M_{c(\text{eff})} = \frac{aM_A + bM_B - \frac{(X + E)(aM_A + bM_B)}{^{3/2}(2a - X - Y) + [(X + E)/2]}}{^{3/2}(2a - X - Y) - ^{3/2}(X + E)} \quad (1)$$

This equation differs slightly from the equation in reference (1) owing to an error in the original calculation of secondary amine reacted, which should be $2a - X - Y$. An improved correction for dangling ends ($X + E$) is used above rather than the previous X . The net result is that the calculated M_c values are higher, principally for samples with 60–100% excess amine. The term on the right side of the numerator in the equation, which is included to subtract out the molecular weight of the dangling ends, implies that they have the same molecular weight as the average segment in the system, i.e., the system molecular weight divided by the total number of segments.

The above procedure has given results which agree within experimental error with both swelling and dynamic mechanical measurements of M_c .³

The design of adhesive-bonded joints involves selection of the proper geometry, consideration of the adhesive and substrates to be employed, the size and dimensions of the joint, and the ease by which it can be fabricated. The simple lap joint is the most commonly used for test purposes; its principle virtues are ease of preparation and loading. It is well known, however, that the distribution of stresses in a lap joint is quite complicated and that the relationship between the failing load of such a joint and the true bond strength between the adhesive and the substrate cannot be determined with confidence. The main objective in the present work was to investigate the bond strength in terms of resin network structure, so that a system which was relatively free of stress concentrations was more desirable. As far as our objective was concerned, the so called "napkin ring" type of joint (first suggested by de Bruyne, 1951) seemed better than the other types of joints.¹⁰ Pure shear and nearly uniform stress field are attained by use of a thin-walled tubular butt joint in torsion. In the elastic range, a material under shear experiences no change in volume, so that no constraint effects occur. A direct relation between torque and shear stress and a state of uniform shear are approached by using a cylindrical joint if the joint width is small compared with the radius. The joint design used in the present work will be described in detail in the experimental section.

Most metals exposed to air are covered with oxide films, which can be associated with widely varying amounts of water, either in the form of adsorbed molecules or as hydrated oxide. The necessity for detailed instruc-

tions on surface preparation indicates that the surface oxide film plays an important part in the adhesive bonding of aluminum and aluminum alloys. There are a number of oxides of aluminum which possess widely different structures and densities, and their properties have been reviewed.⁴

EXPERIMENTAL

The adhesive joint used in the present work is shown in Figure 1 below. Features of this design were found to be a significant improvement over standard techniques. Deformation in the metal thin-walled tube must be avoided, since twisting of the metal tube will cause axial stress components in the joint. In the present design, the tube deflection and thus axial stresses were minimized through use of a very short tube, 0.050 in. in length. Even at this length and a joint thickness of 0.010 in., about 5% of the total deflection of the assembly occurred in the metal ring. It was found that the contact area on the "ring" side of the joint was essentially the same as on the "flat" side and that adhesive failures occurred randomly, with no preference for one side or the other. In early tests with this design, about 40-50% of the joints contained gaps. This was found to be due to gas expansion inside the annular ring during curing, and a small vent hole in each joint raised the percentage of joints without gaps to above 90%.

The torsional test of the joint was conducted on an Instron Universal Testing Machine, Model TM-S (registered trade mark of Instron Engineering Co.). Torque was applied to the joint via a torsion device (based on a design by A. E. Carden) manufactured by Instron as part of the Instralab

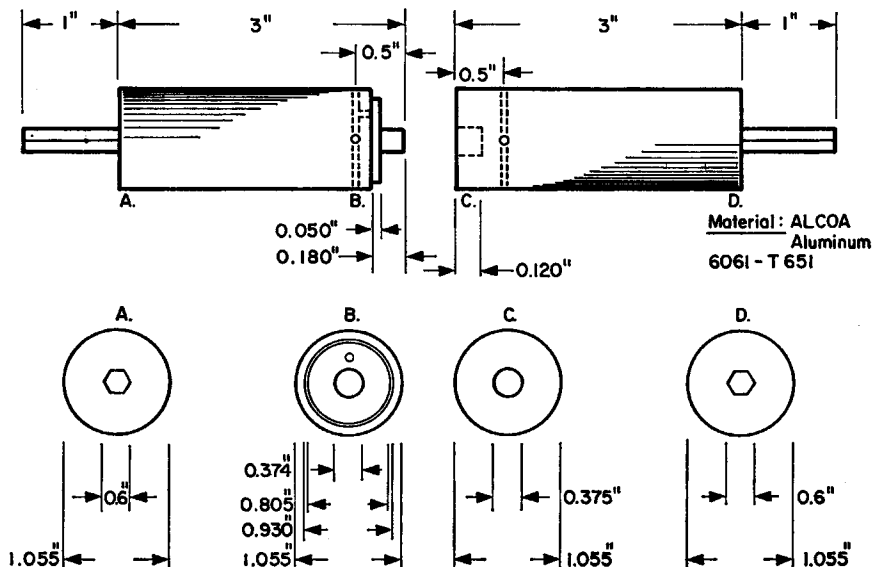


Fig. 1. Aluminum joint assembly.

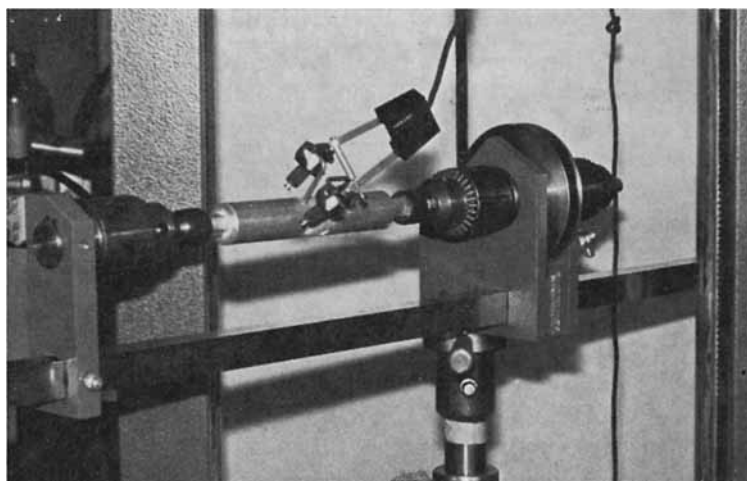


Fig. 2. Instralab torsion apparatus.

series. This device, shown mounted in Figure 2, converts the vertical cross-head motion to a torsional mode by means of a strap wound on a low-friction pulley. The device was used satisfactorily to approximately 450 lb force on the strap after the strap had been changed to 0.012 in. thickness from the standard 0.007 in. thickness.

An external X-Y recorder was used to accept the signals from both the strain gauge signal conditioner (angle of twist signal) and the Instron load cell amplifier. The force applied to the torsional pulley and the deflection of the strain gauge extensometer were therefore obtained from the X-Y recorder readings.

The relationship between the applied torque M_t and the maximum shear stress which it produces is shown below⁶:

$$\tau_{\max} = \frac{16 \cdot M_t \cdot D_o}{\pi \cdot (D_o^4 - D_i^4)}$$

where D_o and D_i are the outer and inner diameters of the ring of resin, respectively. The angle of twist is

$$\theta = \frac{32 \cdot M_t \cdot L}{\pi (D_o^4 - D_i^4) \cdot G}$$

where G is the modulus of rigidity of the ring of resin and L is the resin thickness.

The fact that we have a short projection on a rigid rod rather than a hollow tube is important. Bryant and Dukes⁷ tested longer hollow tubes, but in that case the metal may have deformed, causing axial stresses in the resin and thereby lower strengths and lower strength reproducibility.

Aluminum Surface Pretreatment

The aluminum joints were machined before each reuse with a lathe. The freshly machined joints were polished on a 12 in.-diameter polishing wheel using alumina (suspension in distilled water) of particle sizes 0.5 μ , 0.3 μ , and 0.05 μ , respectively.

After polishing, the surface was cleaned with running tap water, followed by acetone and air dried. The joints were placed in a resin kettle which had a reflux condenser attached, and were then degreased with trichloroethylene vapor for about $\frac{1}{2}$ hr. After the joints were cool, they were submerged for 10 min in a 150–155°F solution of 10 parts by weight of sulfuric acid (sp. gr. 1.84 at 60°F) to 1 part of sodium dichromate to 30 parts of distilled water. The treated surface was flushed with tap water at 150°F and air dried at 150°F for at least 7 hr. The joints were allowed to cool to room temperature before exposure to the resin. The laboratory temperature and humidity were relatively constant at ca. 75°F and ca. 40% R.H., but it is recognized that closer control of time and % R.H. would have been more desirable, based on the results of Wegman.⁵

The joints were then prepared according to the following procedure:

1. The desired amounts of Epon 828 (registered trade mark of Shell Chemical Co. for monomers based on the diglycidyl ether of biphenol A) and methylene dianiline (MDA) were weighed in aluminum dishes, each heated at 100°C, and mixed together for about 3 min.

2. 44 mg (by weighing) of resin were applied on the annulus ring of the joint at room temperature.

3. The two halves of the joint were mated, using an aluminum pipe as sleeve.

4. The joints were placed in an oven with accurate temperature control. The resin was cured at room temperature for 50 min, then 80°C for 1 hr, and finally 150°C for 2 $\frac{1}{2}$ hr.

TABLE I
Sample Preparation and Characterization Data^a

Weight of Epon 828, g	Weight of MDA, g	Excess amine, %	Unreacted epoxy groups, meq/g	Unreacted primary amine groups, meq/g	$M_{c(eff)}$ (mol. wt. between crosslinks, calcd)
12.053	3.123	-0.10	0.125	N.D. ^a	350
21.280	6.098	9.5	0.030	N.D.	360
9.667	3.037	21.2	N.D. ^a	N.D.	420
10.467	3.689	36.0	N.D.	0.03	560
6.899	2.595	45.1	N.D.	0.03	630
10.243	4.270	60.8	N.D.	0.16	820
9.817	4.499	76.8	N.D.	0.24	1250
3.675	4.342	93.0	N.D.	0.48	3200

^a N.D.: Not detectable. Uncured Epon 828 contains approximately 5.25 meq epoxy groups/g. The unreacted epoxy and amine results are per gram of cured resin.

5. The joints were cooled slowly by slightly opening the door of the oven.

The above procedure resulted in samples in which the curing reaction was essentially complete, as evidenced by epoxy or amine group disappearance. Typical residual epoxy and primary amine values are given in Table I.

Torsional Test Reproducibility

The experimental data showing the reproducibility of the test for two different aluminum surface preparation procedures are listed in Tables II and III. A comparison with the reproducibility of various joint tests by other workers is shown in Table IV. One observes that the procedure used in this work gave very good reproducibility in comparison with the other testing techniques. Admittedly, the technique of placing the proper

TABLE II
Torsional Test Reproducibility. Joints in which the Aluminum Surfaces were Polished and Vapor Degreased, but not Chemically Treated*

Force to break, lb	Max. shear stress, psi	Force to break, lb	Max. shear stress, psi
240	7010	221	6460
235	6860'	235	6860
233	6810	228	6660
241	7040	240	7010
		Ave. 234	Ave. 6840
		S.D. ^b = 7, or	S.D. ^b = 199, or
		3% of mean	3% of mean

* An approximately stoichiometric ratio of Epon 828 and MDA was used, and the cross-head speed was 0.05 in./min.

^b S.D.: Standard deviation

TABLE III
Torsional Test Reproducibility. Joints in which Aluminum Surfaces were Polished, Vapor Degreased, and Chemically Treated with Sulfuric Acid-Sodium Dichromate Solution*

Force to break, lb	Max. shear stress, psi	Force to break, lb	Max. shear stress, psi
310	9060	314	9180
306	8950	315	9210
316	9240	308	9010
312	9120	311	9090
308	9010	314	9180
312	9120	310	9060
309	9030	309	9040
312	9120	314	9180
309	9030		
		Ave. 311	Ave. 9096
		S.D. = 2.82, or	S.D. = 81.9 or
		0.9% of mean	0.9% of mean

* An approximately stoichiometric ratio of Epon 828 and MDA was used. The cross-head speed was 0.05 in./min.

TABLE IV
Comparison of the Reproducibility of Various Joint Tests

Type of joint	Strength level, psi	S.D., psi ^a	Worker
Epoxy-Al:lap joints	1510	231 (14%)	R. F. Wegman ⁵
Epoxy-steel:circular butts in simple shear	2840	311 (11%)	Bryant and Dukes ¹⁰
Epoxy-steel:circular butts in torsion	3750	160 (4%)	Bryant and Dukes ¹⁰
Epoxy-steel:tubular butts in torsion	2250	109 (5%)	Bryant and Dukes ¹⁰
Epoxy-Al:modified tubular butts in torsion (no acid-dichromate treatment)	6840	199 (3%)	this work
Epoxy-Al:modified tubular butts in torsion (after acid-dichromate treatment)	9060	83 (1%)	this work

^a The numbers in parentheses are $\frac{\text{standard deviation}}{\text{mean}} \times 100$.

amount of resin on the annular ring requires a delicate touch; one improves with practice.

Measurement of the Shear Strength of the Cured Resins

In order to compare the bond strength of the joint to the shear strength of the resin alone, a punch-type shear strength test apparatus similar to ASTM D-732-46 was used.

RESULTS

Effect of Molecular Weight Between Crosslinks on the Bond Strength of Joints in Which the Aluminum Surfaces were not Chemically Treated

The experimental data for the joints in which the aluminum surfaces were not chemically treated are shown in Table V. Each value reported is the average of three determinations. All of the joints failed adhesively, i.e., the failure occurred at the resin-metal interface.

There appeared to be no ordered effect of the different amine concentrations (and therefore different M_c) on the bond strength. The breaking forces for 0.05 in./min cross-head speed were between 250 and 290 lb. For the joints without chemical treatment, the bond strengths were lower than when chemically treated surfaces were used.

The experimental data for the joints with the aluminum surfaces treated chemically are shown in Table VI. The failure of the joints was a mixture of both cohesive and adhesive types, i.e., neither 100% cohesive failure nor 100% adhesive failure. The average bond strength decreases as the molecular weight between crosslinks increases, for both 0.05 in./min and

TABLE V
Experimental Data for Joints in which the Aluminum Surfaces were
Machined, Polished, and Vapor Degreased, but not Chemically Treated.

Excess amine, %	Cross-head speed, ^a in./min	Force to break, lbs	Ave. shear stress, psi
2.3	0.05	252	6870
	0.20	275	7500
21.0	0.05	265	7230
	0.20	275	7500
50.8	0.05	257	7010
	0.20	282	7690
76.5	0.05	271	7390
	0.20	292	7960
92.9	0.05	250	6820
	0.20	290	7910

^a A speed of 0.05 in./min corresponds to a strain rate of 0.4%/min; 0.20 in./min corresponds to a strain rate of 1.6%/min.

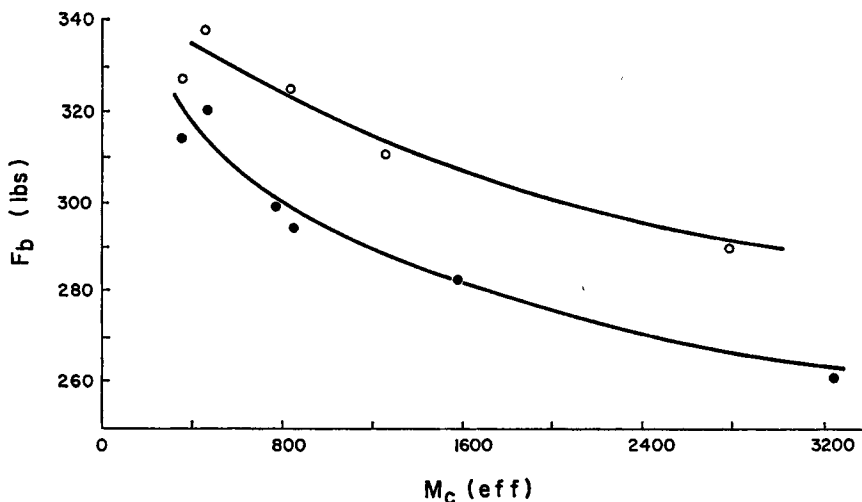


Fig. 3. Relation between M_c and force to break. Chemically treated joints: (●) 0.4%/min strain rate, (○) 1.6%/min strain rate.

0.2 in./min cross-head speeds. A plot of the breaking force versus $M_{c(\text{eff})}$ is given in Figure 3. A 15–20% decrease in bond strength is observed as M_c increases from 330 to 3300.

Effect of Testing Speed

Experimental data showing the bond strengths for various testing speeds are given in Table VII. The bond strength increases toward a limiting value as the testing speed increases from 0.02 in./min to 0.8 in./min. It appears that the limiting value will be reached at a higher rate for the higher M_c (91% excess amine) samples.

TABLE VI
Experimental Data for Joints in which the Aluminum Surfaces were
Machined, Polished, Vapor Degreased, and Chemically Treated^a

Excess amine, %	$M_{c(\text{eff})}$	F_b^+ (Ave.), lb	Max. shear stress, psi	G^*_E , psi $\times 10^{-4}$	G^{**}_E , psi $\times 10^{-4}$
0.05 in./min Cross-Head Speed (0.4%/min Strain Rate)					
1.3	350	314	9180	13.1	13.1
21.1	420	325	9500	13.2	13.1
49.2	750	299	8750	14.1	14.2
60.8	820	293	8570	14.2	14.5
76.8	1250	286	8360	14.7	14.6
80.3	1540	282	8250	14.6	14.6
93.1	3230	259	7560	15.7	15.7
0.20 in./min Cross-Head Speed (1.6%/min Strain Rate)					
1.9	360	327	9560	14.6	
21.2	420	338	9880	15.0	
60.8	820	326	9530	16.1	
76.8	1250	310	9060	16.8	
91.4	2740	292	8540	17.0	

^a G^*_E : Modulus of rigidity of the resin calculated from the torsion of the joint;
 G^{**}_E : Modulus of rigidity of the resin calculated from Young's modulus of F_b^+ ; force to break.

TABLE VII
Effect of Cross-Head Speed on the Bond Strength of Joints in which the
Aluminum Surfaces were Machined, Polished, Vapor Degreased, and
Chemically Treated

Excess amine, %	Cross-head speed, in./min	Strain rate, %/min	$F_b(\text{ave.}), \text{ lb}$	Ave. shear stress, psi	G_E , psi $\times 10^{-4}$
1.3	0.05	0.4	314	8560	13.0
2.0	0.20	1.6	327	8920	14.6
3.2	0.40	3.2	333	9080	15.3
2.0	0.80	6.4	327	8920	15.4
91.4	0.20	0.16	248	6760	14.1
93.0	0.05	0.4	259	7060	15.7
91.3	0.10	0.8	284	7740	16.3
91.3	0.20	1.6	292	7960	17.0
91.1	0.40	3.2	311	8480	17.4
91.1	0.80	6.4	322	8780	17.5

Shear Strength of the Cured Resin (Nonbonded)

The experimental data for the shear strengths of the cured resins are shown in Table VIII. The shear strength is not appreciably affected by the distance between crosslinks, except perhaps at the highest % excess amine levels.

By comparing the shear strength of the resin with the shear strength of a joint, one can infer whether the joint failed adhesively or cohesively. It is clear that for the joints with the aluminum surfaces treated chemically

TABLE VIII
Shear Strengths of Cured Resins^a

Excess amine, %	Shear strength, psi	Standard deviation, psi
-3.70	8490	360 (4.2%) ^b
19.6	8790	346 (3.9%)
51.7	8390	125 (1.5%)
70.2	8530	290 (3.4%)
81.3	8200	297 (3.6%)
92.9	7510	335 (4.5%)

^a A punch-type of shear test was used at 0.05 in./min Instron cross-head speed. Values reported are the averages of 4-5 determinations.

^b Numbers in parenthesis are the standard deviation/mean $\times 100$.

(Table VI), the bond strength almost equals the shear strength of the resin, and many of the failures can be considered as cohesive.

The modulus of rigidity of the cured resin derived from the stress-strain curves agrees very well with the theoretical value calculated from Young's modulus of a similar series of resin samples.⁸ Data are shown in Table VI. Poisson's ratio for the resin was assumed to be 0.34.⁹ The formula

$$E = 2G(1 + \mu)$$

was used, where E is Young's modulus from the stress-strain curve, μ is Poisson's ratio, and G is the shear modulus of rigidity. The speed of testing was 0.05 in./min.

When two materials of dissimilar modulus (aluminum, 10.3×10^6 psi; Epon 828-MDA, 0.4×10^6 psi) are linked together in series, the behavior of the assembly will be controlled by the modulus of the weaker material. If the difference between the two materials is great, the modulus of the assembly should approach the modulus of the weaker material.

Effect of Unreacted Epoxy Endgroups

A few experiments were conducted to determine if unreacted epoxy or amine groups contribute to the interfacial bonding. A series of bonds containing from 0 to 56% excess (over the stoichiometric amount) of epoxy groups was prepared. The results of breaking these bonds are shown in Table IX. From these data it is concluded that excess epoxy groups per se have little effect on the bond strength in the high-strength range. Unfortunately, one cannot draw conclusions about the effect of excess epoxy

TABLE IX
Effect of Excess Epoxy Groups on Bond Strength

Excess epoxy, %	Force to break, lb
0	314
32.9	316
56	311

groups on the lower-strength, adhesive failures. Further studies are in progress.

We can infer from Figure 3 that excess amine groups do not help the adhesion, since the bond strength decreases as the excess amine and residual amine content increases.

Mechanism of Bonding

There are strong *indications* that chemical bonding occurred in the case of joints in which the aluminum surface was treated chemically, although additional work is needed for verification. For the chemically treated surface, the adhered epoxy resin did not come off from the surface after refluxing for several hours in pyridine (a good solvent for the epoxy resin). The epoxy resin came off very easily from surfaces which were not chemically treated. As shown in Table X, when pure Epon 828 was heated with chemically treated aluminum dust, the epoxy content of Epon 828 changed from original 5.23 meq/g to 4.63 meq/g. This change in epoxy content is a preliminary result and needs to be confirmed by further work.

TABLE X
Analysis for Epoxy Groups After Heating with Aluminum
Powder (Simulated Cure Cycle)

Decrease in number of epoxy groups, meq/g. resin		
After heating of Epon 828 only	After heating Epon 828 with untreated Al dust	After heating Epon 828 with chemically treated Al dust ^a
No change from original 5.23 meq/g	0.19	0.67
	0.28	0.64
	0.27	0.75
	Ave. 0.25	0.72
		ave. 0.60

^a Concentration of acid-dichromate bath was 0.1 of the concentration used for the joints. After the cure cycle and epoxy analysis, the samples were mixed with CCl₄, and the aluminum dust was filtered out. An infrared spectrum on the CCl₄ solution could not be distinguished from the spectrum of pure Epon 828; there was no indication of formation of ether linkages or other epoxy reaction products.

CONCLUSIONS

Effect of Molecular Weight Between Crosslinks. From the experimental data, the average bond strength decreases as the molecular weight between crosslinks increases for the chemically treated aluminum joints. For the joints without chemical treatment, the molecular weight between crosslinks seems to not significantly affect the bond strength.

The bond strengths of joints that failed cohesively, or in a mixed cohesive-adhesive mode, are very high. This is because the resin strength is high. The shear strengths which were obtained correspond to tensile

strengths of 11,500–12,000 psi. If such strengths can be obtained reproducibly, epoxy resin–aluminum joints can readily be used for load-bearing applications. The key to reproducibility seems to lie in further study of the mechanism of bonding. If chemical bonding can be verified and the factors which control it optimized, then reliable joints of high strength are a very real possibility. Further work along these lines is under way at this laboratory.

Effect of Surface Preparation. Maximum bond strength was obtained for the joints with the aluminum surface machined, polished, vapor degreased, and chemically treated. For the joints with the aluminum surface machined, polished, vapor degreased, and without chemical treatment, the bond strength was about 80% of the maximum bond strength obtained with chemical treatment. For the joints with the aluminum surface degreased only, the bond strength was only about 65% of the maximum bond strength.

Effect of Testing Speed. The bond strength increased to a limiting value as the test speed increased from 0.02 in./min to 0.8 in./min (or the strain rate increased from 0.16%/min to 6.4%/min). The limiting value appears to occur at higher testing rates as the molecular weight between crosslinks increases.

Effect of Test Method. The torsional test method used in this work appears to have advantages over previous methods. The annular ring is short and rigid, minimizing deformation of the metal fixture and thereby resulting in excellent reproducibility. Since failure is in simple shear, the results from this type of test have wider applicability than results from butt or lap joints, which are dependent on the geometry of the joint.

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