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NOTE

EFFECT OF HEATING ON THE STRUCTURE OF AN ADHESIVE JOINT, AS INDICATED BY ELECTRICAL RESISTANCE MEASUREMENT

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Heating from 20°C to temperatures as low as 25°C was found to cause a partly reversible effect on the structure of an adhesive (epoxy) joint involving steel adjoining (bonded) components with a surface roughness of 120 μm, as shown by monitoring the contact electrical resistivity of the joint during heating and cooling. The reversible portion was due to thermal expansion of the adhesive. The irreversible portion was due to an irreversible microstructural change in the adhesive. These structural changes decreased the extent of electrical contact between the adjoining surfaces at asperities, thereby increasing the contact resistivity.

Keywords: Joint; Adhesive; Electrical resistance; Heating; Steel; Epoxy

INTRODUCTION

Adhesion [1–4] is a commonly used method of joining [5]. Compared with other forms of joining, such as welding, brazing, soldering, and fastening, adhesion tends to suffer from the degradation of the joint at elevated temperatures [6] and in water [7].

The thermal degradation of an adhesive joint is partly due to that of the adhesive, which is a polymer, whether thermosetting or

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thermoplastic. Upon heating past the glass transition temperature, softening occurs.

Another concern relates to the thermal expansion of the adhesive upon heating, as a polymer tends to have a higher value of the coefficient of thermal expansion (CTE) than a metal. In case the adjoining components have CTE values that are quite different from that of the adhesive, the thermally induced stress involved varies with temperature and may cause thermal degradation of the joint.

Of fundamental interest to the understanding of the strength of an adhesive joint is the structure of the joint [8]. Aspects of the structure include the porosity in the adhesive, the asperities on the surfaces of the adjoining components, the thickness of the adhesive, the proximity between the adjoining surfaces at asperities, the roughness of the adjoining surfaces, the extent of asperity deformation, interfacial reactions, etc. The effect of heating on the structure of an adhesive joint sheds light on the mechanisms of thermal degradation.

Previous work on the structure of an adhesive joint involved microscopy [9, 10] dielectric measurement [11–13], ultrasonic methods [14–17], mechanical testing [18, 19], and neutron radiography [20, 21]. These methods tend to be limited in sensitivity to minor structural changes. Some of the methods are destructive and some are not suitable for real-time monitoring. In contrast, this work uses electrical resistance measurement, which gives information on the extent of electrical contact of the adjoining electrically conductive surfaces at asperities and is suitable for monitoring in real time, say, during temperature variation. Real-time monitoring allows observation of the structural changes during heating and cooling, and during thermal cycling, thus enabling distinction between reversible and irreversible effects. In contrast, observation after the fact (rather than during heating and cooling) allows observation of only irreversible effects.

Steel is the most widely used engineering material, so this study uses steel as both of the adjoining components. Epoxies are among the most widely used adhesives, so an epoxy is used in this study. Steel-adhesive-steel adhesive joints are encountered in automotive [15] and other industrial applications.

EXPERIMENTAL METHODS

The steel used was cold rolled 1010, in the form of a sheet of thickness 1.10 ± 0.02 mm. For the purpose of control of the surface roughness, the steel sheet was sand blasted (average particle size of sand 120 μm, 100 grit, Black Beauty Sand, Reed Minerals, Highland, Indiana, USA). The steel sheet was cut into strips of length 63.5 mm and width

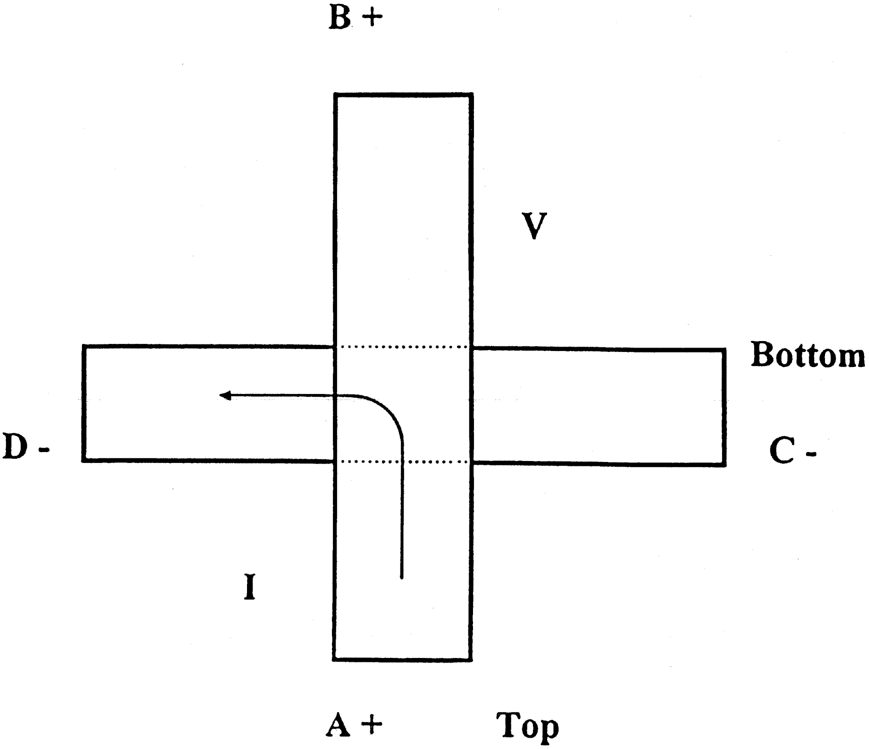

FIGURE 1 Specimen configuration. A, B, C, and D are the four electrical contacts. A and D are for passing a current I. B and C are for voltage V measurement.

5.0 mm. Two strips of steel were positioned to cross one another at 90°, with one strip on top of the other, as shown in Figure 1. Epoxy (nonconductive, 2-part, maximum operating temperature = 107°C, E-Pox-E 5, Dura a Loctite Corp., Sydney, Australia) was applied at the 5.0×5.0 mm square interface of the steel strips. Curing of the epoxy was allowed to occur for at least 3 days at room temperature and a constant pressure (compressive stress in the direction perpendicular to the interface) ranging from 0 to 1.91 MPa. The pressure was provided by known weights and was varied for the purpose of investigating the possible effect of curing pressure on the joint. With the asperities on the steel neglected, the thickness of the epoxy after curing was 0.15 ± 0.06 mm. The thickness of the sandwich was 2.30 ± 0.03 mm. All thicknesses were measured by cross-sectional optical microscopy.

Electrical resistance measurement involved measurement of the contact electrical resistivity of the joint using the four-probe method [22]. This method involves the use of four electrical contacts on the specimen. The outer two contacts are for passing a current. The inner two contacts are for voltage measurement. The advantage of this method is that the contact resistance is not included in the measured resistance between the inner contacts. In contrast, the two-probe method involves two contacts, both of which are for passing a current and for voltage measurement. The two-probe method suffers from the inclusion of the contact resistance in the measured resistance.

The four electrical contacts (A, B, C and D in Figure 1) were in the form of silver paint applied to the end regions (length = 7.6 mm) of the four legs. Current (DC) was passed from A to D while the voltage between B and C was measured. A Keithley 2001 multimeter (Keithley Instruments, Inc., Cleveland, Ohio, USA) was used. The voltage divided by the current was the contact resistance. The contact resistivity was the product of the contact resistance and the contact area ($25.0\,\mathrm{mm}^2$). The contact resistivity is independent of the contact area and is the quantity used in this work to characterize the structure of the joint. The greater the extent of contact between the adjoining steel surfaces at the asperities, the lower the contact resistivity.

For investigation of the effect of heating using an external heater (powered by a 10 V DC power supply), the contact resistivity was continuously measured while the temperature was cycled from room temperature up to about 45°C by using a small resistance heater for heating and using compressed air and a copper tubing with flowing water for cooling. The temperature was measured by using a Type-T thermocouple placed at the top surface of the junction shown in Figure 1. The heating and cooling rates are as shown by the plots of temperature versus time in Figures 2–4.

RESULTS AND DISCUSSION

Figures 2–4 show the effect of initial heating and cooling on the contact resistivity. In every thermal cycle the resistivity increased, due at least partly, to the thermal expansion of the adhesive upon heating and the consequent decrease in the extent of electrical contact of the adjoining steel surfaces. With the asperities of average height 120 μm (according to the average particle size of the sand used in sand blasting) and the epoxy thickness 150 μm, even a small thermal expansion (0.2 μm, as calculated for a temperature increase of 15°C, under the assumption that the CTE of epoxy is 100×10^{-6}/°C [23])

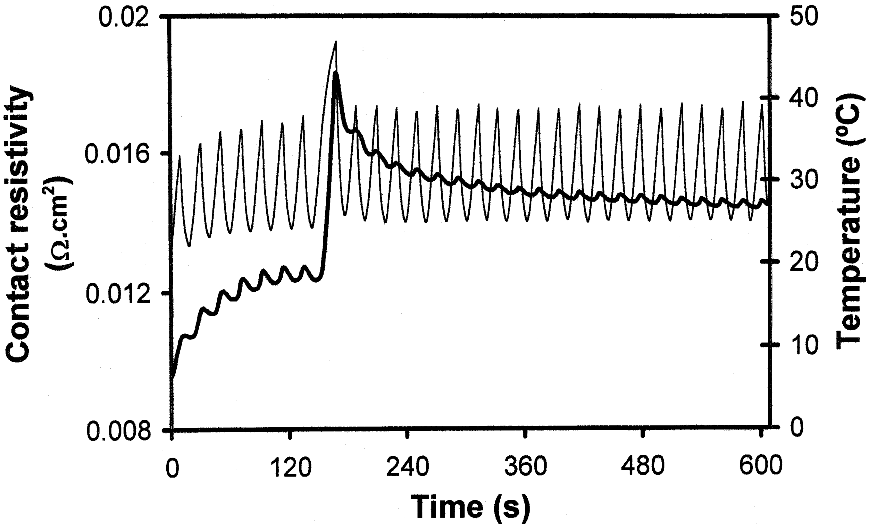

FIGURE 2 Variation of the contact resistivity with time and of the temperature with time during thermal cycling with an abrupt increase in the amplitude of temperature variation. The curing pressure was 1.9 MPa. Thick line, resistivity; thin line, temperature.

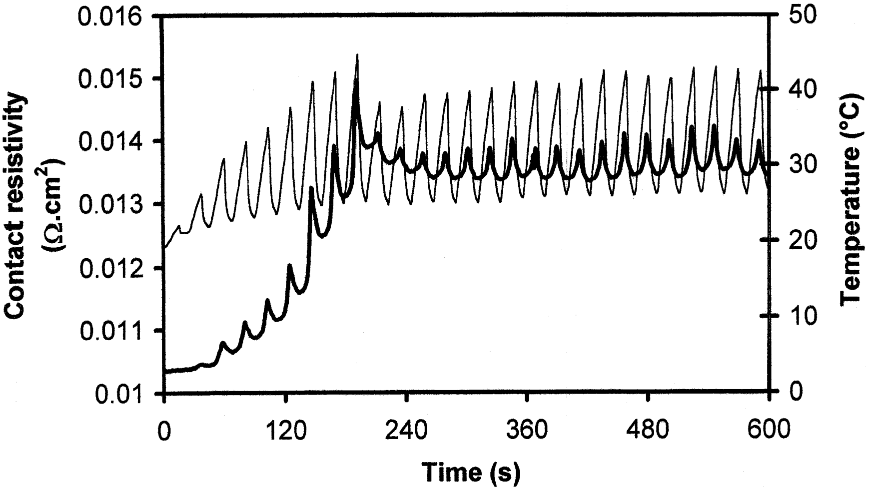

FIGURE 3 Variation of the contact resistivity with time and of the temperature with time during thermal cycling with a gradual increase in the amplitude of temperature variation in the first 9 cycles. The curing pressure was 1.0 MPa. Thick line, resistivity; thin line, temperature.

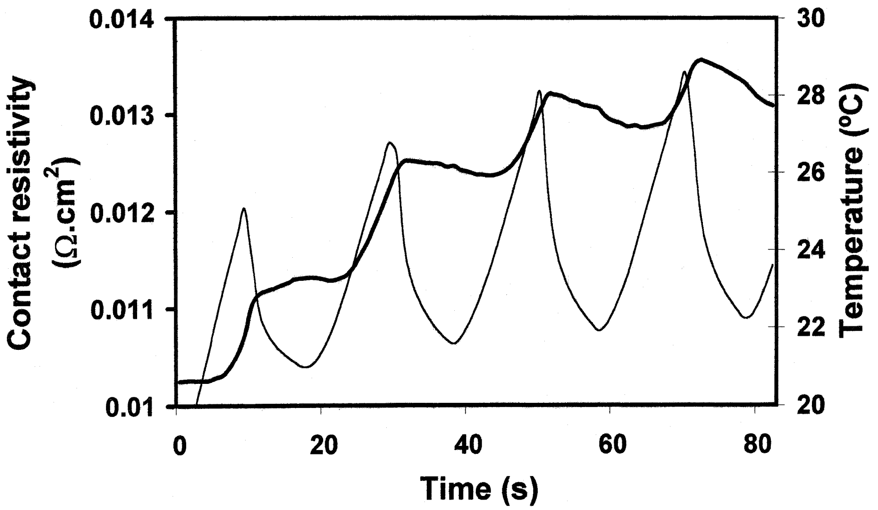

FIGURE 4 Variation of the contact resistivity with time and of the temperature with time during thermal cycling with a gradual increase in the amplitude of temperature variation in the first 4 cycles. The curing pressure was 1.9 MPa. Thick line, resistivity; thin line, temperature.

may cause an observable increase in the contact resistivity. However, the resistivity increase may also be due to a microstructural change at the interface between epoxy and steel. The greater the temperature increase, the larger was the increase in contact resistivity, as shown in the initial 9 thermal cycles in Figure 3.

A sudden increase in the amplitude of temperature variation caused an abrupt increase in the contact resistivity, as shown in the eighth cycle in Figure 2. The abrupt resistivity increase was largely irreversible, although the extent of partial reversibility increased with time upon subsequent cycling (Figure 2). In other words, the abrupt resistivity increase was accompanied by an abrupt upshift of the resistivity baseline. Before the abrupt increase in the amplitude of temperature variation, the resistivity baseline upshifted gradually from cycle to cycle (Figure 2). However, the extent of upshift was small compared with that resulting from the abrupt increase in the amplitude of temperature variation.

When the amplitude of temperature variation was increased gradually, as in Figure 3, the resistivity baseline shifted upward cycle by cycle. An expanded view of the initial few cycles shows that the resistivity increased with increasing temperature, even when the maximum temperature in a cycle was only 25°C (first cycle in Figure 4).

The observed resistivity effects were essentially not affected by the curing pressure used in the adhesion process, in contrast to the decrease in resistivity upon increase in pressure for a metal-particle-filled adhesive [24]. This is probably due to the relatively small distance between the metal particles in a conductive adhesive and the consequent increased sensitivity to pressure.

The irreversible portion of the observed resistivity increase is attributed to a microstructural change, which decreased the extent of electrical contact of the adjoining surfaces. In contrast, the reversible portion is attributed to thermal expansion. The observed resistivity increase upon heating was partly reversible. This means that the observed resistivity increase was partly due to thermal expansion and partly due to a microstructural change. In spite of the low magnitude of temperature variation, with the maximum temperature as low as about 25°C (Figure 4), partial irreversibility was observed. At the sites where the adjoining surfaces almost touched each other through asperities, the microstructural change may diminish the proximity between the adjoining surfaces at asperities and/or (although less likely) affect the ease of electron transport across the epoxy. The microstructural change probably impaired the electron transport across the epoxy from one steel component to the other. It is possible that this microstructural change is related to the degradation of the interface between epoxy and steel as the epoxy underwent additional cross-linking upon heating. However, the nature of the microstructural change has not been identified. The transport can occur via tunneling and/or electric field mechanisms.

This article shows that reversible and irreversible changes occur in an epoxy joint upon heating to temperatures as low as 25°C, even though the maximum operating temperature according to the manufacturer is 107°C. The irreversible effect is of possible concern to the joint quality, although the correlation between electrical and mechanical properties is beyond the scope of this paper.

Electrical resistance measurement was effective for monitoring the interfacial structure of an adhesive joint involving electrically conductive adjoining components (*e.g.*, steel [15, 25] and aluminum [26]) in real time, provided that the adhesive thickness was small enough to allow electrical contact (not necessarily direct contact) between the adjoining components at asperities. The rougher the surfaces of the adjoining components, the larger would be the minimum allowed adhesive thickness for the electrical resistance method to be applicable. The minimum adhesive thickness is approximately equal to the height of the asperities.

CONCLUSION

Upon heating (from 20°C up to a temperature as low as 25°C) an epoxy joint involving steel and adjoining components with a surface roughness of 120 μm, the contact electrical resistivity of the joint increased, with partial reversibility upon subsequent cooling. The reversible portion was due to thermal expansion of the epoxy. The irreversible portion of the observed resistivity increase is attributed to a microstructural change, which decreased the extent of electrical contact (*i.e.*, the number of contact points or the total area of the contact points) of the adjoining surfaces.

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