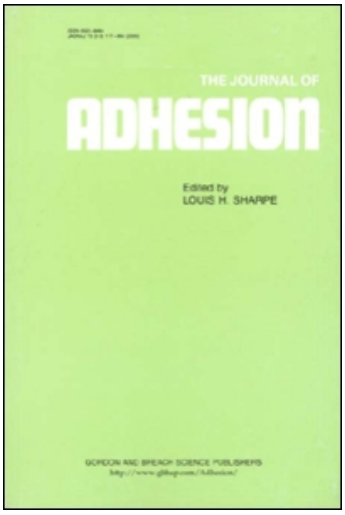

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Scanning Acoustic Microscopy of the Cellular Structure of the Interphase in a Metal-Adhesive Bond

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NOTE

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KEY WORDS Adhesive bond; adhesives; scanning acoustic microscopy; morphology; interphase; imaging.

INTRODUCTION

In the sixties Bikerman[1] deduced from his experiences in adhesive bonding that the adherence between an inorganic solid surface and a polymeric adhesive occured through a "weak boundary layer."

Since then, the concept has evolved among adhesion scientists and it is now accpeted that the adhesive bonding consists of an interphase[2,3] which is three dimensional.[4]

Though experimental evidences are scarce, some recent experiments confirm that idea. Schonhorn[5] has shown that polyethylene has a different structure in contact with gold than it has in the bulk. Cuthrell[6] has observed colloidal structure of epoxy resins solidified in contact with a solid mould. Kötting[7] has obtained microscopic pictures of the columnar structure of an epoxy bond to aluminium. Possart[8] has proved the existence of charges and one of us[9] has explained the conduction through an adhesive joint as the junction of columnar structures initiating from each part of the bond.

It is certainly important to obtain more information on the nature and structure

of the interphases, and on the depth to which the columnar structure exists, in order to understand the adhesion of polymers with inorganic materials.

We have used a scanning acoustic microscope to study the interphase between a metal and polymeric material. The scanning acoustic microscope, compared with other microscopes, has the additional advantage of allowing the observation of both the surface and subsurface features in a nondestructive way. This would help us in studying the difference between the material close to the surface and the bulk material.

SCANNING ACOUSTIC MICROSCOPE

The working principle of an acoustic microscope has been dealt in detail in the literature.[10,11] In short, the heart of the acoustic microscope is a single-crystal sapphire lens. It has at one end a spherical cavity, and at the other end a piezoelectric transducer. Since we are dealing with acoustic waves a coupling fluid like water is used, between the lens and the sample, to propagate and focus them on the sample. A radio frequency is applied to the transducer. The generated ultrasonic waves pass down the sapphire rod, are converted into spherical waves by the cavity, propagate through the coupling fluid, and converge to a narrow waist at the focal plane, where the sample is placed (Figure 1a). A part of the acoustic energy is transmitted into the sample, and a part is reflected back. This reflected energy is picked up by the lens and transducer. The intensity of the reflected signal is plotted as a function of position of the lens over the sample. To obtain an acoustic image the lens is mechanically scanned over the sample.

The resolution of an acoustic microscope, defined as the smallest distance (h) resolved between two points is given by $h = 1.13\ (F/D)\ \lambda$. Here λ is the acoustic wavelength in water, F is the focal length and D is the diameter of the lens. At a frequency of 1.3 GHz, choosing optimal F and D, a resolution of $\sim 0.85\ \mu$m, comparable to that of an optical microscope, has been achieved.

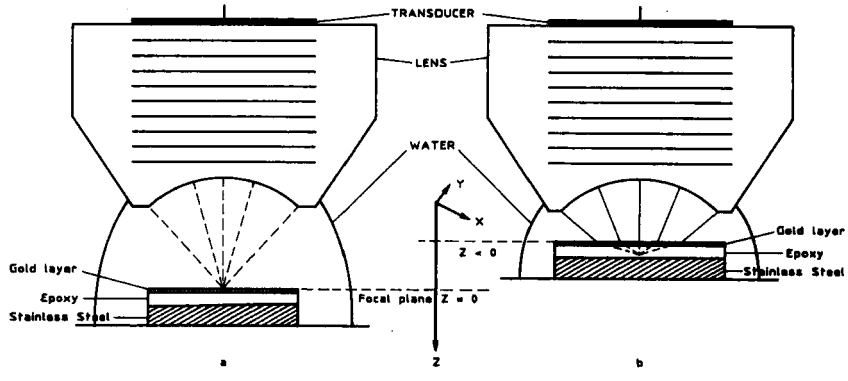

FIGURE 1 Principle of the acoustic microscope: (a) acoustic waves focused on the surface ($Z = 0$), (b) acoustic waves focused in the interphase between gold and polymer ($Z < 0$) (not to scale).

The main source of contrast in an acoustic microscope is the variation of the elastic properties in the specimen. In polymeric materials, when the focus is on the surface (Figure 1a), the contrast could be directly related to the acoustic impedance, which is the product of density and velocity of sound in the material. When the distance between the lens and sample is reduced (defocus, Figure 1b), it may be possible to bring the acoustic waves into focus inside the sample. In this case the subsurface features could be the source of contrast. Since acoustic waves undergo a refraction at the water/polymer interface, the actual defocus is not the same as the real depth of the focal plane inside the sample.

PREPARATION OF SAMPLES

In order to obtain a metallic substrate thin enough to allow the observation of the interphase, even at high frequencies, a layer of gold (purity 99.99, Metalor) 1000 Å in thickness was deposited on a flat sapphire plate (roughness less than 10 Å). We did not deposit any intermediate layer to have low adherence between the gold and sapphire. Then an adhesive bond was made between the gold plate sapphire and the stainless steel plate. The dimensions of both the plates were 4 cm × 1 cm. After polymerisation of the adhesive, the sapphire plate was removed, keeping the adhesive between the stainless steel plate and the gold layer. Care was taken not to damage the gold layer.

ADHESIVES

Two reactive adhesives were considered, A) a low-temperature-curing, two-component adhesive made of DGEBA (828 Shell) and Jeffamine 230 (Texaco); (cure 4 h at 60°C); B) a mono-component, rubber-modified epoxy, E18, similar to CIBA AV 118 (cure 45 min at 160°C).

MICROSCOPIC OBSERVATIONS

In our investigations we have used an ELSAM Leitz (Germany) scanning acoustic microscope. A part of it is an optical microscope. The area to be imaged was chosen optically, so as to be free of visible defects, and imaged acoustically. This procedure was followed to avoid the dominance of contrast due to defects formed during sample preparation.

Optical images are simply bright without any visible structure and hence are not shown here. Acoustical images have been obtained with a lens of half opening angle 50° and working at a frequency of 1.3 GHz, which have a resolution of 0.85 μm.

RESULTS AND DISCUSSIONS

Sample A

The acoustical images, when the focal plane is exactly on the surface of the sample, usually show very little contrast as in the optical images. But as the

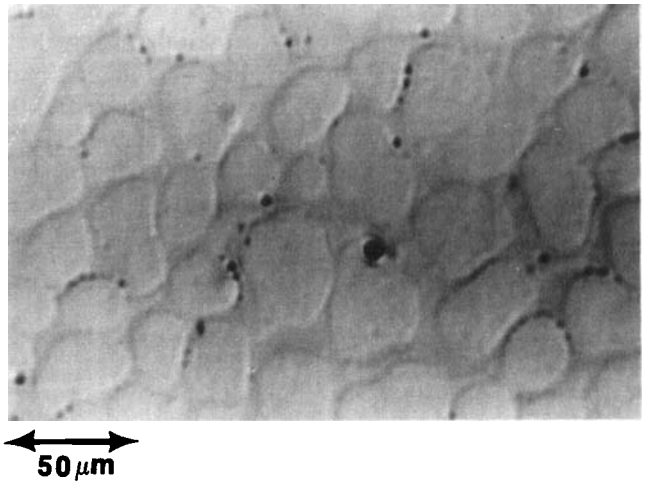

FIGURE 2 Acoustic image of the interphase in sample A, [DGEBA (828 Shell) and Jeffamine 230 (Texaco), (cure 4 h at 60°C)] for an acoustic wave of 1.3 GHz, (resolution 0.85 μm), at a defocus $Z = -2\,\mu$m.

defocus is increased, a cellular structure becomes clear. In Figure 2 we show an image taken at a defocus of $Z = -2\,\mu$m, where the cellular structure is very clear. As the defocus is increased further to $Z = -4\,\mu$m this structure disappears. The planar size of the cells is in the range of 15–30 μm. Imaging was done on three different places, and images were similar; therefore, in Figure 2 we have shown only one example.

Sample B

An acoustic image taken at a defocus of $Z = -2\,\mu$m is shown in Figure 3. Here also we can see clearly the cellular structure. As in the case of sample A, an image at a defocus of $Z = -4\,\mu$m does not show the cellular structure. The size and shape of the cells are similar to that of sample A.

In both the low temperature (60°C) and high temperature (160°C) curing adhesives a cellular structure, with similar features, has been observed. This structure only exists in the vicinity of the gold/adhesive adhesive interphase. In the bulk the structure is different. Since the elastic properties of these adhesives are not known precisely, the depth up to which this structure exists cannot be determined accurately. An approximate value of 3–4 μm can be deduced from the ratio of the velocity of sound in water to the velocity of sound in the adhesive, and the defocus ($Z = -4\,\mu$m) at which the substructure disappears. Velocity of sound in the adhesives is calculated using a modulus of 2400–3600 MPa and density of 1100 to 1230 $\mathrm{kgm^{-3}}$ given in a handbook[12] for similar adhesives. A velocity of 1500 $\mathrm{ms^{-1}}$ has been used for water.

No apparent changes in the cell morphology have been observed as a function of defocus.

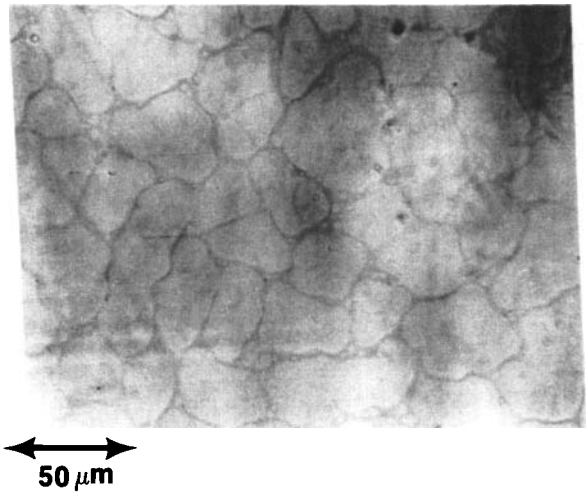

FIGURE 3 Acoustic image of the interphase in sample B, [rubber modified epoxy E18 (cure 45 min., at 160°C)] for an acoustic wave of 1.3 GHz, (resolution 0.85 μm), at a defocus $Z = -2\ \mu$m.

Summary

Scanning acoustic microscopy provides clear evidence for a cellular structure at the gold/adhesive interface. Flocules of 15 to 30 μm are observed over a depth of a few microns in the vicinity of the gold layers. These cells recall the observations of Cuthrell[6] and may explain the columnar structure observations of Kötting.[7]

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