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ULTRASONIC WAVE INTERACTIONS WITH INTERFACES*

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We have now heard about why we need a method of measuring the bond strength and Dr. Tennyson Smith has just showed us what the bond line looks like on a microscopic scale. Obviously the strength measuring method must deal with very small dimensions and a very complicated series of different layers. It boils down to a pretty tall order to consider how an ultrasonic test is going to examine these very thin layers and diffuse interfaces to give a viable test method.

The objective of the work that I will describe was to determine experimentally how a sound wave interacts with a layer whose thickness is measured in atomic sized units. That is, the interface layers are measured in hundreds of angstroms and are thus much, much thinner than any of the millimeter or tenths of millimeter kinds of wave lengths that we ordinarily have, or can hope to have to interrogate the bond line.

The question we must address is; How does a very long wave length ultrasonic wave interact with a very thin layer or collection of layers? Or; When we get a signal back from an interface, how should we unfold it to learn something about the nature of the interface?

There are some crude models available that describe how sound waves go through layers, and we shall begin by considering these in order to develop some ideas on what to look for in data sets. Unfortunately, this approach immediately brings us face to face with a serious problem that clouds any attempt to study an adhesive bond with ultrasonic waves. This problem is that the adhesive bonds with which we wish to work lie between a metal and some polymeric adhesive -- for example, a bond line between an adhesive layer and aluminum.

If one sends a sound wave through the aluminum to the bond line, it will be reflected by the large impedance discontinuity that exists between the metal and the polymer so that the information in the reflected wave will be dominated by the discontinuity in impedance instead of the effects of thin interface structures.

The experiments I will describe were undertaken to try to get rid of this acoustic mismatch reflection so that I could observe only what happened right at the thin bond line itself.

To accomplish this I utilized a suggestion of Dave Kaelble who pointed out that two identical pieces of Lucite or polystyrene can be bonded together in different ways to provide a set of specimens having the same kind of material on each side of the interface, but with different bond strengths at the interface.

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In this way the sound wave reflected back from the interface would be characteristic of sound reflected from a very thin layer and not from a gross impedance mismatch.

There are two ways of making a bond between two pieces of Lucite; one chemical and the other, thermal. The chemical method is probably familiar to all of you who have stuck pieces of plastic together. It consists of purchasing a solvent that is a liquid to be coated on the surface of the two parts. This chemical dissolves a thin surface layer and makes it soft so that when the two parts are pressed together, a good intimate contact between the two surfaces is achieved. The semi-liquid nature of the interface allows the molecules from one block to crosslink with those of the other to provide a solid bond when the solvent diffuses away from the interface. The other method of making a chemical bond between plastic specimens is to take the two blocks of plastic to a temperature that is above the glass transition temperature. Here again, the interface material is made soft to allow an external pressure to provide intimate contact while thermal agitation of the molecules provides the crosslinking to form the bond line. By controlling the time, temperature and pressure of this kind of bond, it is possible to make bonds of different strengths.

We were successful in making a series of both chemical and thermal bonds of different strengths. For our chemical bonds, we used a mixture of methylene chloride and ethylene chloride on Lucite blocks. Since the two blocks of Lucite were large, we were able to observe slightly different acoustic reflections from different regions indicating that different regions had different acoustic characteristics. After the acoustic data had been recorded, we cut the specimens up, fabricated tensile test bars and observed that the different regions actually showed somewhat different strengths. Another specimen was prepared with a commercial adhesive at its interface and it exhibited both a different acoustic response and strength.

The thermal treatment specimens were prepared by subjecting Lucite blocks to a pressure of 40 Kg/cm² for 15 minutes at a series of different temperatures. At the completion of the ultrasonic tests, we fractured these specimens and observed a nice gradation in strength that corresponded to the temperature at which the joint had been made. The results of the strength measurements on the specimens prepared in the different ways are summarized in Table I.

Our ultrasonic test plan was to take sound waves in the convenient range of from 1 to 10 Megahertz, shine them at normal incidence onto the interface and measure the reflection that returns as a function of frequency in order to generate data on the frequency dependence of the reflectivity of interface.

Dr. Dick Elsley of the Science Center programmed our computer to perform a Fourier analysis of the signals reflected from the interface. This method not only yielded the reflected amplitude as a function of frequency, but also the phase shift due to the reflection as a function of frequency.
Table I.

Mechanical strengths and ultrasonic reflection coefficients (relative to an air interface) observed for adhesive bonds formed in different ways between Lucite blocks.

<table>
<thead>
<tr>
<th>BONDING METHOD</th>
<th>MEAN BOND STRENGTH kg/cm²</th>
<th>CHARACTERISTIC ULTRASONIC REFLECTION STRENGTH dB</th>
</tr>
</thead>
<tbody>
<tr>
<td>CHEMICAL:</td>
<td></td>
<td></td>
</tr>
<tr>
<td>60% METHYLENE CHLORIDE 40% ETHYLENE DICHLORIDE</td>
<td></td>
<td></td>
</tr>
<tr>
<td>REGION A</td>
<td>364 ± 36</td>
<td>-10.5</td>
</tr>
<tr>
<td>C</td>
<td>383 ± 39</td>
<td>-14.5</td>
</tr>
<tr>
<td>B</td>
<td>409 ± 24</td>
<td>-25.0</td>
</tr>
<tr>
<td>ACRYLIC ADHESIVE PS-30</td>
<td>94 ± 16</td>
<td>-12.5</td>
</tr>
<tr>
<td>THERMAL:</td>
<td></td>
<td></td>
</tr>
<tr>
<td>120°C, 15 MIN.</td>
<td>0 - 10</td>
<td>-48.0</td>
</tr>
<tr>
<td>130°C, 15 MIN.</td>
<td>17</td>
<td>-48.0</td>
</tr>
<tr>
<td>140°C, 15 MIN.</td>
<td>76 ± 7</td>
<td>-48.0</td>
</tr>
<tr>
<td>160°C, 15 MIN.</td>
<td>69 ± 9</td>
<td>-48.0</td>
</tr>
<tr>
<td>190°C, 15 MIN.</td>
<td>310 ± 50</td>
<td>-48.0</td>
</tr>
<tr>
<td>200°C, 15 MIN.</td>
<td>408 ± 30</td>
<td>-48.0</td>
</tr>
</tbody>
</table>
Before we look at the ultrasonic measurements themselves, it is instructive to go to the literature and try to predict what we might expect to see in terms of the reflected amplitude and phase as a function of frequency. Three models for how a thin interface reflects sound waves can be considered. These are described qualitatively by the diagrams shown in Fig. 1 where I have drawn the reflected amplitude as measured in db below a reflection from an air to Lucite interface as a function of frequency. The horizontal axis is the thickness of the layer divided by wavelength, which is the same as frequency but is a more physically significant parameter. At the top is shown the good old classical thin sandwich layer model which shows the very characteristic reflection amplitude that goes to a maximum when the thickness is one quarter wavelength, and to a minimum when the thickness is one half wavelength. This classical case describes the situation of a sharp discontinuity in impedance between medium 1 and medium 2. If we consider the case of the impedance fuzzed out to give a gradual variation in impedance with distance, the reflected amplitude can be calculated in closed form only for certain mathematical forms for the shape of the impedance-distance curve. We used one of these special forms to get a qualitative idea of what the reflection coefficient as a function of frequency should look like. The middle graph in Fig. 1 shows that the reflection coefficient has the same general shape as before with a peaked reflection near a one fourth wavelength thickness and zero (that was at 1/2 of a wavelength) moved out to higher frequencies. Actually, for some choices of thickness and impedance mismatch there is no zero in reflectivity at all. A third interface model available in the literature treats the thin interface as a discontinuity in displacement across a plane over which the stress is continuous. If we're going to go to thin interfaces, we're going to be looking in detail at the frequency dependence down here in the thin region where the reflection coefficient rises as a function of frequency. But for thicker bonds we may be able to see some maximum to get information on the thickness of the bond line.

This model is appealing because it represents the case of a bond line that has become infinitesimally thin having no thickness at all. It is then better mathematically to talk about it in terms of the boundary conditions at an interface instead of as a layer. Two authors have worked out how a sound wave reflects from an interface across which the stress is continuous, but the displacement undergoes a discontinuous jump whose magnitude is proportional to the stress. If you wish, you can imagine such an interface as a collection of little springs that have a certain spring constant, and yet mathematically are of infinitesimal length. In this case, we get a reflection coefficient that rises with frequency very much like the variation observed in the thin limit of the finite layer problems discussed above.

Examination of Fig. 1 shows that all of our models yield a frequency dependence of the reflection that rises with increasing frequency and may go through a maximum if there is a real layer whose thickness is comparable to the wavelength.

Returning to our experimental measurements in which we used digital techniques and Fourier analysis to deduce both the amplitude and the phase
Fig. 1. Schematic diagrams of the frequency dependence of the reflected amplitude of ultrasonic waves reflected from the various impedance versus distance functions shown on the right.
as a function of frequency it was anticipated that the phase should do a lot of interesting things, especially in cases of a layer of 1/2 wave length thickness. In order to make sure that the computer and everything was working correctly, we carefully studied the classical case of a well defined layer (top case of Fig. 1) formed by two steel blocks with a layer of water in between. By adjusting the thickness of the water layer so that at 6 megahertz it was 1/2 wave length thick, we observed the big dip in the reflection coefficient predicted by the theory as well as the predicted phase variation. These data as displayed by the computer driven plotter are shown in Fig. 2. The phase shift goes through the expected variation at the frequency of the reflectivity minimum although it does not quite make it from the plus 90 to minus 90 degree phase shift predicted by the theory and the reflectivity does not really get completely down to 0.

However, the data are quite adequate to demonstrate that our Fourier analysis procedure is working satisfactorily. In order to further check our methods of measurement in the limit of a very thin layer we made a sample in which a thin sheet of gold leaf was sandwiched tightly between two plastic blocks. By weighing a known area of the gold leaf prior to bonding, we deduced that it was only 2000 angstroms thick. (I didn’t believe it when I measured it so I had some of the other fellows do it, and they got the same answer.) Intimate contact between the gold and the plastic was achieved by pressing the sandwich together at an elevated temperature in a vacuum. The resulting specimen consisted of a thin planar layer having well defined acoustic properties embedded in a plastic medium also with well defined elastic constants. The very small reflection coefficient of this layer was measured with narrow band transducers operating at 2 1/4, 5 and 10 MHz to define the frequency dependence of the reflection for comparison with theory. The solid line shown in Fig. 3 is the theoretical prediction for a layer with sharp boundaries and a thickness of 2600 Angstroms. Thus our measurement technique works both quantitatively and qualitatively.

Application of the Fourier analysis technique to the chemically bonded plastic blocks yielded the reflected amplitude versus frequency functions displayed in Fig. 4. Obviously, the frequency characteristics are peaked functions characteristic of well defined layers with rather sharply defined boundaries. The height of the peak measures the degree of acoustic impedance mismatch between the solvent rich medium inside the layer and the plastic surrounding the layer. Also indicated on the graphs is the strength of the bond for these three in the solid curves measured in subsequent destructive tests. It can be seen that the impedance mismatch gets smaller and smaller as higher strength bonds are achieved. Furthermore, there is a tendency for the peak to shift to higher frequencies with increasing strength indicating that the layer has gotten a little thinner. These results are gratifying because it indicates that for chemically formed bonds it is possible for ultrasonics to determine the thickness of the bond line as well as the strength. The dashed curve describes the data obtained on the bond formed by a commercially available adhesive. Its strength is different because it has a different chemical composition and one would probably have to work out exactly what the relationship between strength and reflected amplitude maximum is for each type of chemical used for forming the bond.
Fig. 2. Frequency dependence of the amplitude and phase of the ultrasonic signal reflected from a thin layer of water held between parallel steel blocks.
Fig. 3. Frequency dependence of the amplitude of ultrasonic signal reflected from a 2000 Angstrom thick gold film between two blocks of Lucite. The data points were obtained at three fixed frequencies and are in dB relative to reflection from a Lucite to air interface.
Fig. 4. Graphs of the frequency dependence of the ultrasonic energy reflected from adhesive bonds between Lucite blocks formed by placing an adhesive or solvent at the interface. These data were deduced from Fourier transforms of the RF signal reflected from the bond line.
Figure 4 demonstrates that ultrasound is able to tell what the bond strength and thickness are for these chemically formed layered bonds. Obviously, these results are somewhat fortuitous because the half wavelength thicknesses of the layers are approximately 0.1 millimeter and hence cause strong, frequency dependent responses in the frequency range below 10 MHz. Such bonds are, of course, much thicker than those anticipated at the adhesive to aluminum interface where the important layers are a few hundred Angstroms thick. Therefore, the thermal bonds made by squeezing two pieces of plastic together were investigated by reflecting sound waves from the interfaces. Table I shows that a series of specimens having different strengths were available. Figure 5 shows the results obtained from the digital, Fourier analysis of the ultrasonic reflection from three specimens of quite different strength. The reflected amplitude as a function of frequency shows that there is actually no correlation with the strength and that the reflection coefficient is not terribly frequency dependent. Since the interface reflections were small, very careful measurements of the reflected amplitude at three fixed frequencies (2 1/4, 5 and 10 MHz) were made just to make sure the computer results were correct. These data are displayed in Fig. 6 which shows that the reflected levels lie between 45 and 50 db below an air interface and that there is still no correlation between reflected amplitude and the strength. Each shape of data point corresponds to a different bond strength and there is no real order to the location of these points.

Furthermore, all of the data lie in a horizontal band across the figure indicating that the reflection efficiency is pretty frequency independent. Unfortunately, all of the mathematical models are very frequency dependent. By forcing a fit at 5 MHz, the theoretical curve for a well defined thin layer misses both the low and the high frequency points. By adjusting the parameters in the diffuse layer model, it is possible to achieve some degree of frequency independence but again the theoretical curve falls off too fast at low frequencies. Thus the diffuse layer model doesn't fit either. Of course, the boundary condition model gives a rising function which doesn't fit at all.

Our conclusion is that these thermally made bonds, where we anticipate a very thin interface, reflect sound waves at normal incidence in such a way that there is no correlation with strength. This leads to two possible explanations. Either looking at the bond line with normal incidence is too insensitive to the strength property, or in this particular experiment there was some artifact at that interface that produced a frequency independent scattering, that dominated the reflection from the true chemical interface in which we were really interested. These interface artifacts might be dust particles or scratches or some thin thing left at the interface after bonding. It will be necessary to do some further experiments to find out what is taking place.

It is interesting to note that we observe a frequency independent reflection from the thin interface. Analysis of many scattering models shows that one must twist the parameters all out of shape to get such a result. I must therefore conclude that we were not able to get a good
Fig. 5. Frequency dependence of the amplitude of ultrasonic signals reflected from three bonds between Lucite blocks whose failure strengths are indicated. These data were deduced from a Fourier transform of a broad band ultrasonic pulse.
Fig. 6. Comparison between various theoretical models for the frequency dependence of the reflection of ultrasonic energy by a planar impedance variation with measurements made on a bond between Lucite blocks formed at the different temperatures indicated.
mathematical model to fit the data that were observed. The data are correct but we must go back and do some more work and look at some different kinds of models for reflection at interfaces. I also think that the results represent a warning that we should start looking right away at other kinds of ultrasonic interactions with the interface, and in particular, waves that propagate along the interface or reflect from the interface at large angles. In short, we will have to be a little more subtle with our sound waves.
DISCUSSION

DR. LACKMAN (Rockwell International, B-1 Division): We have time for a few questions.

DR. WOLFGANG SACHSE (Cornell University): George, what did you mean by reflection amplitude or the thing you plotted on the vertical axis. What is that quantity?

DR. ALERS: We measured the peak to peak voltage amplitude of the echo reflected from an air interface to define a standard, frequency independent signal level. Then, we did the same for the echo reflected from the interface in question and took 20 times the logarithm of the voltage ratio. In actual practice we simply added gain in decibels to bring the amplitude of the interface reflection back up to the same voltage deflection on the oscilloscope that was observed for the amplitude reflected from an air interface.

DR. SACHSE: Then, you made essentially two measurements, but did you have different bonds between the first measurement and the second measurement?

DR. ALERS: Yes, we had different bonds but we found that by repeating the same bonding techniques we had adequately reproducible bonds.

DR. HENRY BERTONI (Polytechnical Institute of New York): When you were modeling this, did you introduce any loss in the medium?

DR. ALERS: No, we're waiting for Joe Rose to tell us what horrible effects the losses will cause.

DR. SY FRIEDMAN (Naval Ship Research and Development): In your model, you assumed that the interface layer had a higher acoustic impedance than the base medium. Would it not be just as valid to assume that it had a lower acoustic impedance in which case you might have gotten quite a different theoretical calculation?

DR. ALERS: That's a good point. For the bond with sharp sides it doesn't make any difference. The ratio of the impedance, whether it's greater or less than 1, comes in squared and you get the same features. Now, for the diffuse bond it makes a big difference whether you're talking soft or hard material inside the interface. However, it turns out that when you go to the limit of a very small impedance mismatch in the diffuse layer case as we experienced here, you couldn't tell the difference. They both, whether it was hard or soft, gave the same frequency dependence at 40 db down below the air interface reflection. In the case of the discontinuity, the sign of that $k$ factor in front of the stress, that describes the compliance at the interface comes in squared also and hence doesn't affect the reflection coefficient.

MR. ALEX GARY (NASA-Lewis): Is it possible that the fact that you applied pressure for the thermal bonding made a difference?
DR. ALERS: Oh, yes. We worried about that and put other specimens into the furnace and pushed on them to give them the same thermal treatment. They came back with about the same properties as specimens that hadn't been through the process. So, our standard specimen could be one that had been around the lab all the time or one that had been through the same heat treatment.

PROFESSOR HARRY TIERSTEN (Rensselaer Polytechnical Institute): In the thin case, how did you estimate k?

DR. ALERS: I didn't ever have to estimate k.

PROFESSOR TIERSTEN: You never used that?

DR. ALERS: It didn't fit. It gives a rising frequency dependence that did not fit the experimental data. In the initial plan of the experiment I was curious to find out what k would be, but I never got any data that ran along the predicted curve.

PROFESSOR TIERSTEN: I don't think there should be such a relationship.

DR. LACKMAN: We have time for one more question.

PROFESSOR YIH PAO (Cornell University): That calculated dip of the reflection coefficient, don't you see a repeat pattern for higher frequency?

DR. ALERS: Yes

PROFESSOR PAO: Have you measured that experimentally?

DR. ALERS: Oh, sure.

PROFESSOR PAO: You only showed the one dip, but it would go on for higher frequencies?

DR. ALERS: Yes. If I made my layer thicker, I could get lots more dips in it. Yes, they are there but I just didn't bother to show all of them. In the diffuse layer case the dips are more complex. If the material in the interface is harder, one gets a whole lot of those dips. If the layer material is softer, those dips move to higher frequencies and one can actually lose them entirely. So, there's a qualitative difference in the diffuse layer case.