BOND STRENGTH MEASUREMENTS BY ULTRASONIC SPECTROSCOPY

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ABSTRACT

The goal of this project has been to discover techniques for predicting the strength of a metal-to-metal adhesive bond from nondestructive measurements on the completed structure. Both the cohesive strength of the adhesive material itself and the adhesive strength of the metal-to-adhesive interface must be determined separately. In previous phases of the program, it was demonstrated that the Fourier transform of the ultrasonic echo returned from a metal-adhesive-metal sandwich structure immersed in a water bath contained sufficient information to obtain a prediction of the cohesive strength of the joint. Furthermore, certain features of the Fourier transform were shifted by thin layers of "different" materials at the metal-to-adhesive interfaces so that detecting poor adhesion was also a possibility. During the current phase of the program, more reliable mechanical tests and more accurate measurement techniques were developed. As a result, measurements of the wave velocity in FM-400 adhesive joints subjected to different degrees of cure correlated with the cohesive shear strength of the joints. Quantitative measurements of the standing wave resonant frequencies in Chemlok 304 adhesive joints showed a correlation with the strength of adhesion at the metal-to-adhesive interface.

Introduction

The nondestructive measurement of the strength of a completed adhesive bond between two pieces of metal is a major problem which is retarding the application of this very efficient method of joining to aerospace and other lightweight structures. Without a method of verifying the strength of a finished joint, the designer, the manufacturer, and the user are reluctant to utilize adhesive bonding for primary structures and thus heavy, complicated joints made with holes and bolts are still the dominant construction method. Over the past several years, various NDE techniques have been applied to this problem, and considerable success has been achieved at locating regions of disbonds or foreign objects whose dimensions are on a scale large compared to the bond line thickness. Such methods as ultrasonics and x-rays or neutron radiography can produce maps of the adhesive material which show these gross flaws in the joint. What is lacking are techniques which expose deficiencies in the intrinsic strength of the joint which arise from poor cohesive strength in the bulk of the adhesive itself or from the lack of adhesion between the adhesive and the metal. Poor cohesion within the adhesive can arise from an improper cure during manufacture, incorrect chemistry in the original adhesive material, or microscopic defects such as porosity. Poor adhesion can arise from incorrect preparation of the metal surfaces prior to bonding, aged adhesive material or an improper primer application to the metal. It has been the objective of this study to find ultrasonic techniques that can give a quantitative measure of the presence of these intrinsic deficiencies and provide a means for predicting the strength of adhesive bonds which do not show gross flaws under conventional radiographic or ultrasonic C-scan inspections.

Accomplishments to date include the development of computer programs to analyze the ultrasonic pulse signals reflected from adhesively bonded sandwiches in such a way that numerical values for the ultrasonic velocity and attenuation within the adhesive layer can be extracted. It was shown that both of these physical properties could be correlated with the strength of bonds made with a model adhesive system. Thus it appears that techniques are available for deducing the cohesive strength of a finished adhesive bond from quantitative measurements of its response to ultrasonic waves. The current program, reported here, has been to demonstrate this strength prediction capability on bonds made with commercial adhesives after being subjected to various environmental exposures and/or various fabrication technique modifications.

The detection and measurement of a reduction in quality of adhesion at the metal to adhesive interface has been a much more challenging problem for ultrasonic inspection. Theoretical investigations concerning the way in which the mathematical boundary conditions at the adhesive to metal interface effect the ultrasonic wave propagation modes in the sandwich structure have shown that standing wave resonances in the thickness dimension are sensitive to the state of the boundary. Therefore, the current program has been to detect these thickness mode resonances with the same type of computerized apparatus that was used in the cohesive bond strength measurements and to see if a correlation with the strength of adhesion could be demonstrated.

Experimental Techniques

In order to establish any quantitative relationship between a nondestructive measurement and

the strength of an adhesive joint, each step in the procedure must be performed in such a way that all uncertainties are minimized. The ultrasonic measurements must be made accurately at the location on the bond where the strength measurement is made. Likewise, the strength measurement must be made in such a way that small, localized defects (such as edge imperfections) do not dominate the mechanical failure process. In all cases, the chemistry and curing of the adhesive as well as the preparation of the metallic surfaces prior to bonding must be made in a uniform, consistent manner so that scatter in the results due to fabrication variations are minimized. Each of these three key elements were carefully developed for the present program and are discussed separately below.

Mechanical Tests

In previous studies, the lap shear specimen shown at the top of Fig. 1 has been used to provide a measure of the adhesive bond strength. Its primary drawback is that a stress concentration occurs along the edge of the bonded joint that is perpendicular to the stress and any weakness in this region is then likely to be the source of failure. Since the ultrasonic measurements on the joint must be made at points away from the edges, the standard lap shear test is unsatisfactory for accurate correlations between strength and NDE measurement, because the ultrasonic data comes from regions of the joint that are probably different from the regions that initiate failure. To overcome this problem, two new test procedures were introduced for the current program. These are the peel test and the compression shear test as shown in Fig. 1. The force necessary to cause a crack to propagate through the exact region where ultrasonic measurements have been made is measured in the peel test. Since the small compression shear test specimen is cut out of a larger bonded plate after a thorough inspection by ultrasonics, the region of failure initiation can be located exactly where precise ultrasonic measure-

Figure 1. Various methods of testing the strength of adhesively bonded joints.

ments have already been made. For nearly all of the test results presented later in this report, the compression shear test was used on small specimens 1" long and 1/2" wide in which the shear was applied to a 1/4" x 1/2" area in the center of the specimen. These test specimens were cut from 1-inch wide by 6-inch long bonded strips of 1/8-inch thick aluminum or from 6" x 6" bonded plates of 1/8-inch thick aluminum which had been interrogated at an array of points by ultrasonic pulse-echo methods.

Adhesive Bond Preparation

Two types of adhesive bonds were examined in this program. The first was designed to demonstrate the ability to measure deficiencies in the cohesive strength of a completed bond which had been made with a commercial adhesive material (FM 400) but cured at incorrect temperatures. This procedure was applied to the 6"x 6" square panels. The second bond type was designed to demonstrate the ability to measure strength reductions caused by poor adhesion between the adhesive and the metal. This latter class of defect was simulated by preceding the bonding process with the placement of a thin layer of contamination on the 6"x 1" aluminum peel test strips in such a way that the thickness of the contamination would vary slightly along the length of the strip in order to insure obtaining a distribution in strength values out of each long specimen. For these adhesion tests a model adhesive system (Chemlok 304) was used with surface contamination produced by wiping stopcock grease on the surface of the aluminum or by controlled precipitation of stearic acid onto the aluminum from a solution of pentane and stearic acid.

Ultrasonic Tests

Improvements in the ultrasonic test techniques were made to increase the accuracy of capturing the ultrasonic wave forms and placing them in the memory of the computer. Further improvements in the mathematical model which describes the reflection of ultrasonic waves by layered media were incorporated into the computer system so that direct comparisons between measurements of the reflected amplitude as a function of frequency could be made with detailed theoretical predictions of this same function. As a result of these changes, small shifts in the position of the resonant frequencies of the sandwich structure could be reliably measured and compared with theoretical expectations.

The existence of an accurate theoretical model for the ultrasonic signals has played a vitally important role in developing quantitative methods for adhesive bond strength determination. Figure 2(a) shows a schematic diagram of the metal-adhesive-metal sandwich immersed in a water bath and subjected to ultrasonic wave interrogation by a transducer mounted above the sandwich and oriented to send acoustic waves perpendicular to the layers. The reflected signals, labeled R_1, R_2, R_3 and R_4 in the figure, return along the normal to the specimen and are detected by the same transducer that launched them. These reflection signals are shown in Fig. 2(b), which demonstrates the importance of using very broad band (short time duration) ultrasonic signals in order to clearly separate the reflection from the top surface of the aluminum (R_1) from the reflection from the top of the aluminum-adhesive
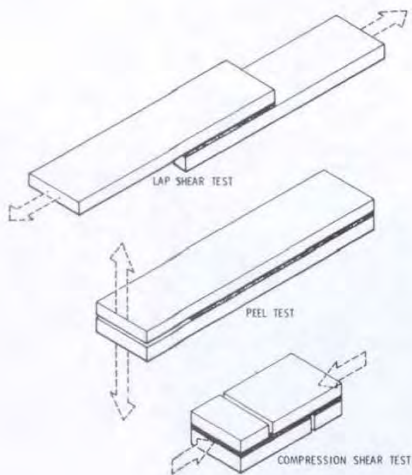
interface (R_2) and from the reflection from the bottom of the adhesive layer (R_3). Figure 2(c) shows an enlarged version of the region of time containing only the reflections from the adhesive layer. This demonstrates that the digital computer analysis system is capable of performing very detailed analyses on specific events in the complex series of echos returned from the total adhesive bond sandwich. In particular, the ratio of the signal amplitudes reflected from opposite sides of the adhesive (the ratio R_3/R_2) is used to measure the ultrasonic attenuation in the adhesive layer. In order to measure the sound velocity in the adhesive layer or the frequencies of standing wave resonances within each of the layers, the computer generates the Fourier transform of the time domain signals shown in Fig. 2 to produce the reflected amplitude versus frequency graph shown in Fig. 3(a). This computation includes correcting the ultrasonic signal for the frequency response of the transducer to produce what is called the Normalized Reflection Spectrum. Because of the finite band pass of the transducer, the computer shows no information below 1 MHz and above 19 MHz, even though there are certainly reflection minima in these frequency ranges. Figure 3(b) shows the results of calculating the reflection spectrum of an aluminum-adhesive-aluminum sandwich with a small amount of attenuation included in the physical properties of only the adhesive. A comparison of the experimental and theoretical spectra demonstrates the high confidence with which various features in the experimental spectrum can be used for quantitative measurements and correlations. The splitting of the individual dips and the relative depths of the two minima are observed in both the theory and the experiments. These prominent dips, spaced at 2 MHz intervals, arise from the standing waves in the aluminum plates and are split into a pair of dips because the plates are of equal thickness and are coupled together by the adhesive layer. Thus their individual thickness resonant modes are shifted upward and downward along the frequency axis depending upon whether the metal surfaces on each side of the adhesive are moving in-phase or out-of-phase. The small dips near 5 and 10 MHz represent the standing wave resonance of the adhesive layer alone and can be used to determine the velocity of sound in the adhesive if the thickness of the layer is known.
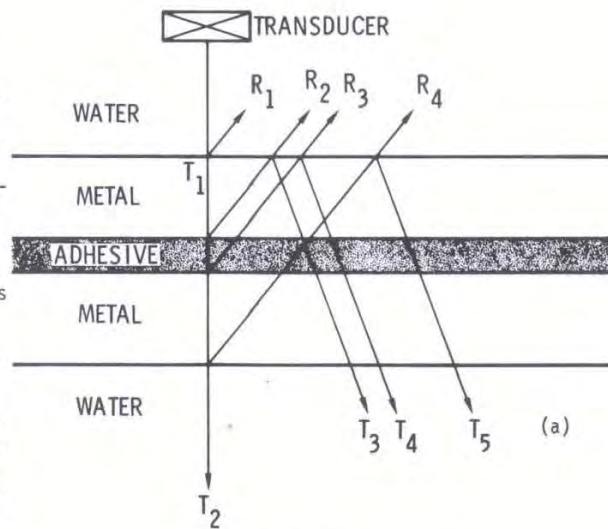
ULTRASONIC INSPECTION
WITH NORMAL INCIDENCE WAVES

Figure 2(a). Schematic diagram showing the position of an ultrasonic transducer over the planar layers that form a common adhesive joint between metal plates.
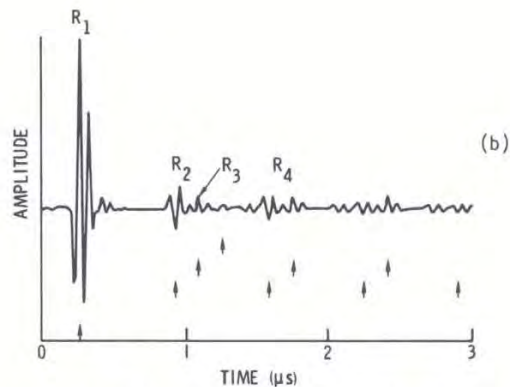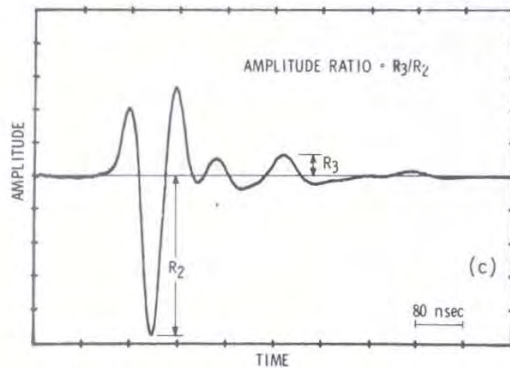

Figure 2(b). Signal amplitude versus time display for the echoes reflected from the multilayered adhesively bonded structure of Fig. 2(a).

AMPLITUDE RATIO = R_3/R_2

TIME DOMAIN PRESENTATIONS OF SIGNALS

Figure 2(c). Expanded view of the time interval in which the reflections from the adhesive layer alone occur.

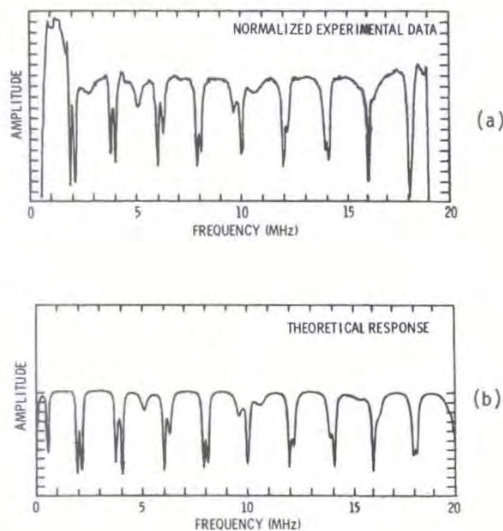NORMALIZED EXPERIMENTAL DATA (a)

THEORETICAL RESPONSE (b)

Figure 3. (a) Reflected amplitude versus frequency graph deduced by the computer taking the Fourier transform of the time domain signal shown in Fig. 2(b). Transducer effects have been normalized out of these data. (b) Theoretical prediction of the reflected signal versus frequency graph based on 1/8-inch thick aluminum adherends joined by a 10 mil adhesive layer with some attenuation in the adhesive.

Cohesive Bond Strength Measurement

Since both the elastic properties and the cohesive strength of the bulk adhesive are related to its internal chemical structure, a correlation between the cohesive strength of the bond and measured values of the velocity of sound and the attenuation of sound in the adhesive layer should exist. Figure 4 shows the good correlation observed in previous studies on a model adhesive system (Chemlok 304) which contained no scrim cloth and which cured without the evolution of any volatile materials. The various values of strength were obtained by changing the ratio of chemical constituants in the precured adhesive.
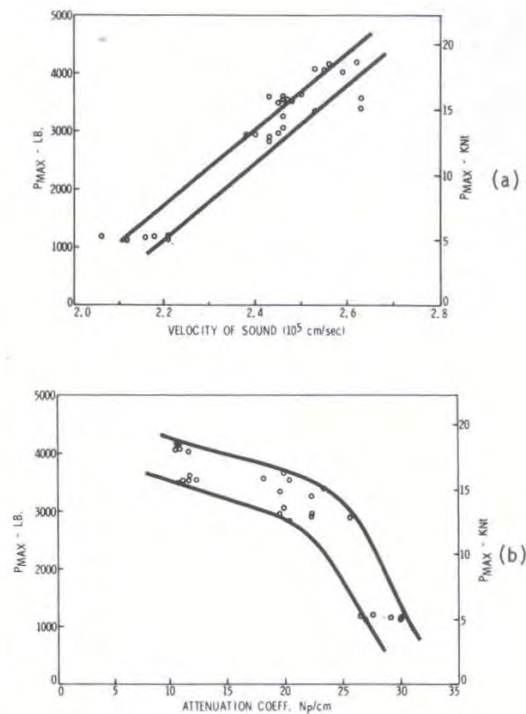
Figure 4. (a) Correlation observed between the velocity of sound in a model adhesive and the strength of the joint as measured by a lap shear test. (b) Correlation observed between the attenuation of sound in the model adhesive and the strength of the joint as measured by a lap shear test.

The current program has been directed at demonstrating this same correlation in a modern structural adhesive (FM 400) which contains both a scrim cloth as well as a metal particle filler and which cures with the evolution of a gas. Variations in the degree of cohesive strength were achieved by performing the cure at different temperatures - a common processing error. Figure 5 shows the data obtained when the ultrasonic measurements of sound velocity and attenuation were plotted against the compression shear strength of the many samples. To obtain these data, two 6"x 6"x 1/8" aluminum plates were bonded together and cured at a particular temperature. Then a computer automated inspection system was used to collect ultrasonic reflection signals and to deduce values for the velocity and attenuation at an array of points over the 36 square inches of bonded surface area. These points were chosen such that when the panel was cut up into a collection of compression shear samples (see the bottom of Fig. 1) the area subjected to the shear stress was identical with that which had been measured acoustically. Obviously the spread in both the acoustic measurements and strength measurements is quite large so that the scatter in the data overshadows any correlation in the case of the attenuation and makes a correlation between strength and velocity rather inaccurate. An examination of the fracture surfaces of the broken samples revealed the presence of scattered arrays of bubbles within the adhesive which had a wide distribution of sizes, although the largest did not exceed the spacing between the fibers of the scrim cloth. Since these bubbles should act as excellent ultrasonic wave scattering centers, they undoubtedly make a major contribution to the measured attenuation and cause it to vary considerably without changing the strength of the bond. Hence, the poor correlation between strength and attenuation. A surprising feature about the relationship between acoustic velocity and strength is the abruptness with which the strength changes as a function of curing temperature. Note that the strength jumps from zero to its maximum value when the cure temperature changes only from 120° C to 140° C. Also note that the values of the velocity corresponding to the uncured and fully cured states extend over the range of 2.5×10^5 cm/sec to 3.2×10^5 cm/sec - a spread of 25%. This spread is comparable to that which was observed with the model adhesive system when its chemical composition was changed and the good correlation between velocity and strength shown in Fig. 4(a) was observed. Thus, the present, real adhesive system differs from the model system only in the degree of uncertainty or scatter in the data. Presumably this scatter is associated with the hetrogeneous nature of commercial adhesives which contain scrim cloth, fillers and bubbles. Further effort should now be directed at understanding the origin of the scatter and correcting for it in the data processing or specimen preparation stages.
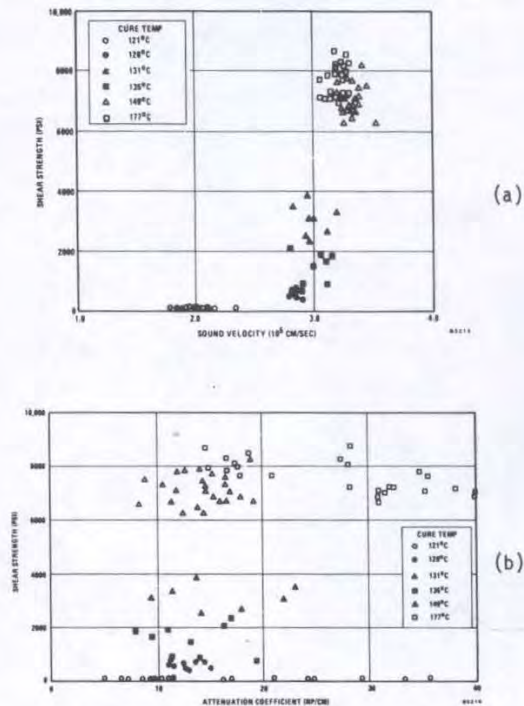

Figure 5. (a) Correlation observed between the velocity of sound and the compression shear strength measured on a joint made with FM 400 adhesive cured at different temperatures. (b) Correlation observed between the attenuation of sound and the compression shear strength measured on a joint made with FM 400 adhesive cured at different temperatures.

Adhesive Bond Strength Measurement

The task directed at a nondestructive measure of adhesion at the adhesive to metal interface concentrated on developing techniques for obtaining a distribution of bond strengths in the geometry of the peel test specimen using contamination on the aluminum adherends to adjust the strength values. It also concentrated on development of very accurate methods for extracting the resonant frequency values from the reflection amplitude versus frequency plots generated by the computer (see Fig. 3). Two methods of contamination were successful. One used a variable thickness layer of stop-cock grease to achieve different lap shear strengths on four specimens. The reflected amplitude versus frequency plots for these four samples are shown in Fig. 6 with the value of the measured shear strength of each sample recorded in the upper right hand corner. Obviously, the highest frequency at which the standing wave resonances are observed to be split increases as the strength of the bond increases. By making a graph of the strength versus the frequency at which the splitting disappears, one gets the excellent correlation shown in Fig. 7, which relates a nondestructive acoustic measurement with the strength of an adhesive bond that fails by weak adhesion. A physical basis for this correlation can be proposed to

arise from the fact that the weaker bonds are asso-
ciated with a thicker layer of grease at the inter-
face and this layer does not pass the high frequency
sound waves from the aluminum into the adhesive.
Thus the high frequency standing wave resonances
show only the standing wave in the upper aluminum
plate because no acoustic energy gets through the
interface to excite the lower aluminum plate at its
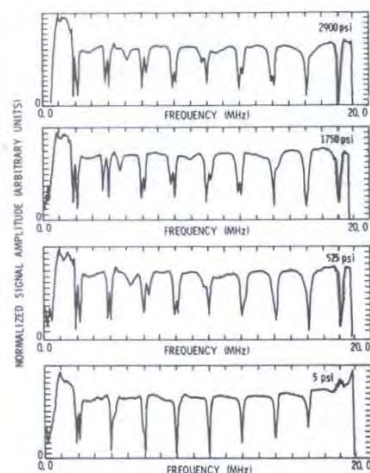higher harmonics.

Figure 6. Reflected amplitude versus frequency
plots measured on four samples whose lap
shear strengths had been reduced by
placing different thin layers of stop-
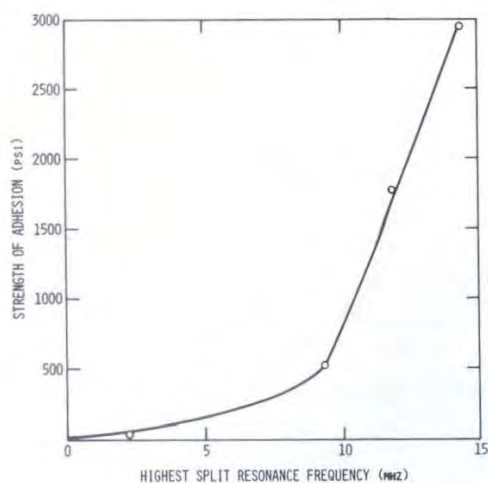cock grease at the adhesive-to-metal
interface.

Figure 7. Correlation between the highest fre-
quency for which a split resonance is
observed and the adhesion strength
observed in a lap shear test.

The other successful method of controlling the
strength of adhesion was to put a variable thickness
layer of stearic acid on the aluminum before prepar-
ing the bonded specimen. By placing a solution of
the acid in pentane on one of the aluminum adherends
and then allowing the pentane to evaporate, it was
possible to deposit a very thin layer of stearic
acid on the aluminum. When the ultrasonic signal
amplitude versus frequency graphs for these speci-
mens were produced, it was observed that the fre-
quency at which splitting of the resonances appeared
did not change when the strength of the bond changed.
Thus the stearic acid layer at the interface does
not show the same ultrasonic filtering response as
the stopcock grease layer exhibited.

During the earlier theoretical studies on the
effects of interfaces on the ultrasonic response of
a sandwich structure, it was predicted that the
lowest standing wave resonance of the total struc-
ture should be particularly sensitive to the condi-
tions at the interface. Preliminary experiments
verified this prediction by showing a strong corre-
lation between strength measurements and the value
of the lowest resonant frequency. To see if this
correlation also existed for interfaces contaminated
with stearic acid, special low frequency transducers
were used to allow the computer system to display
resonances in the frequency range from 0.3 to 0.6
MHz. Because this low frequency resonance is also
sensitive to the thickness of the adhesive layer,
special computer calculations were carried out to
determine how all the standing wave resonances are
shifted by a small change in bond line thickness.
To determine the bond line thickness appropriate to
a given test specimen, the computer system was used
with a high frequency transducer to display the
entire series of high frequency standing waves in-
cluding the resonance of the adhesive layer alone
(shown as the small dip near 5 MHz on Fig. 3). By
assuming that the compliance of the bulk adhesive is
constant over the length of a peel test specimen
with stearic acid at the interface, the frequency of
the adhesive resonance observed at an individual
location can be used to calculate the effective
thickness at that location. This value can then be
used to correct the lowest frequency resonance to
that which it would have for a standard thickness.
In this way, all the lowest frequency resonances for
any set of specimens can be compared to one another
and correlations with other variables such as bond
strength can be investigated. A comparison between
the compression shear strength and the lowest fre-
quency of resonance is shown in Fig. 8(a) for two
specimen sets labeled 271 and 273, and a reasonably
good relationship appears to exist. Figure 6(b)
shows the correlation between the compression shear
strength and the separation in frequency between the
two minima, i.e. the splitting, of the lowest fre-
quency split resonance at 1 MHz in Fig. 3. Here
again, a normalization of the data to constant
effective bondline thickness has been carried out.
This second parameter seems to provide a somewhat
more accurate correlation with strength and a smaller
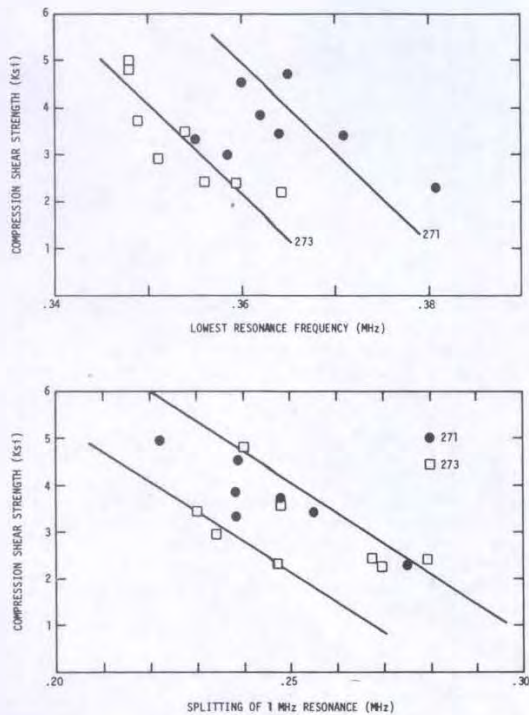spread between the different initial specimens.

Figure 8. Correlation observed between the compression shear strength of two series of samples and two features of their amplitude versus frequency characteristics. Top curve: correlation with the lowest standing wave frequency for the entire sandwich. Bottom curve: correlation with the degree of splitting of the lowest frequency standing wave in one of the adherend plates.

Conclusions

Based on the postulate that the cohesive strength of a layer of adhesive should be related to its bulk physical properties, such as the velocity of sound and the attenuation of sound, a computerized system to rapidly analyze the ultrasonic signals reflected from an adhesively bonded pair of aluminum plates has been assembled. Using this system, it has been demonstrated that a good correlation between the cohesive strength of adhesive joints formed with a model adhesive and its velocity and attenuation can be obtained. On real, commercial adhesives the correlation is not as good because the inhomogeneous nature of this class of materials increases the spread in velocity and attenuation measurements. Methods for correcting the data for these inhomogeneity effects are now being developed.

For the case of measuring the quality of adhesion between the adhesive and the metal, techniques based on making quantitative measurements of the standing wave resonant frequencies of the bonded sandwich structure have been developed. After corrections are made for variations in the thickness of the adhesive bond layer from specimen to specimen, it has been shown that both the lowest standing wave resonant frequency of the entire sandwich and the magnitude of the splitting of the lowest standing wave frequency in the aluminum adherends can be used to predict the strength of the completed bond. Again, considerable scatter in the data points exists and an investigation into the origin of this effect is underway.

Acknowledgement

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