ADHESIVE JOINT EVALUATION BY ULTRASONIC INTERFACE AND LAMB WAVES

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The effectiveness of ultrasonic methods for the evaluation of material properties and process control has been known for a long time. It was also found that ultrasonic methods can be used for material strength prediction and verification. Application of ultrasonic methods for evaluation of adhesive bonds has a number of specific features. It involves checking a thin layer of adhesive, the thickness of which is much smaller than that of adherents. It is necessary to detect not only violation of the continuity of the adhesive, but also its effective elastic properties, which correlate with the strength of the adhesive bonds. Most of the previous techniques used longitudinal ultrasonic waves normally incident on the adhesively bonded interface. But these waves are insensitive to the adhesion properties between adhesive and adherends.

We suggest an alternative method using a guided waves. The advantage of this technique is that this wave produces shear stresses on the interface. This method was previously discussed (ref. (1) - (6)) and reviewed (ref. 7).

In this paper first, we describe application of interface waves for cure monitoring of structural adhesives; second, the Lamb technique which is applicable for study of adhesive joints of thin sheets is discussed.

ELASTIC INTERFACE WAVES

The general case of propagation of interface waves in adhesively bonded interfaces was studied by Rokhlin et al (ref. 1). The case of propagation of interface waves in a system of two half spaces separated by a viscoelastic layer was analyzed. The shear modulus \( \mu_0 \) of the layer was assumed to be smaller than the shear modulus of the half spaces, then in the case of identical half spaces there always exists a guided interface wave.

A possible method for obtaining interface waves is shown schematically in figure 1. The surface wave, excited in the lower substrate, is transformed in the interface region into an interface wave. Due to the closeness of velocities of the surface and interface waves, only a small part of the energy is transformed into bulk waves. The interface wave leaving the interface zone is retransformed into a surface wave, and is sensed by a receiver.

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The deformation of a thin solid interface film by the interface wave is illustrated schematically in figure 2. Figure 2a shows the deformation produced by the surface wave in the subsurface layer. It shows tension and compression regions separated from one another by a half wavelength. Figure 2b shows the deformation of an interface layer, the thickness of which is much smaller than the wavelength. Due to antisymmetry of motion, the interface layer is subjected to shear strain (fig. 2c).

One can estimate the viscoelastic properties of the interface layer by measuring the velocity and attenuation of the interface wave (ref. 1). We showed that the complex shear modulus of an interface wave, \( \mu_0 \), is related by a simple expression to the interface wave velocity \( V_i \).

\[
\frac{\mu_0}{\mu} = \frac{\bar{\eta} \Delta_r}{\beta - \frac{\bar{\eta}}{\rho} \rho_0 a_1 - 2\bar{\eta} a_2 a_2}
\]

where \( \alpha = V_t/V_i \) is the normalized wave number for the interface wave; \( a_1 = \alpha^2 - \beta \gamma; a_2 = 2\beta\gamma - W; W = a^2 + \beta^2; \beta = (\alpha^2 - 1)^{1/2}; \gamma = (\alpha^2 - V_r^2/V_s^2)^{1/2}; \Delta_r = (W^2 - 4\alpha^2\beta\gamma); \Delta_r \) is the characteristic function for the Rayleigh wave, \( V_t \) and \( V_s \) are the shear and longitudinal wave velocities in the substrate, \( \rho_0 \) is the density of the interface film material, \( \rho \) and \( \mu \) are the density and the shear modulus, respectively, of the substrate, \( f \) is the frequency and \( h = 2\pi f h/V_t \) is the nondimensional film thickness, \( 2h \) is the thickness of the film.

Equation (1) is valid at \( h << \chi^0 \), where \( \chi^0 \) is the length of shear waves in the material of the interface film. When this condition is not satisfied, shear modulus \( \mu_0 \), determined from Equation (1) can be refined on the basis of the more exact equations (ref. 1). To determine the complex shear modulus \( \mu_0 \), one should substitute into Equation (1) the complex wave number \( \alpha \), which is calculated on the basis of measured velocity and loss factor of interface waves.
Numerical analysis shows that the sensitivity of this method is high in the range of film thicknesses which are small as compared with the length of interface waves. Hence at film thicknesses from 100 to 1 μm the measurements should be performed at relatively low ultrasonic frequencies from 0.5 to 5.0 MHz.

It is possible to single out the principal features of the method, which indicate that the use of the interface waves for the evaluation of thin interface layers (in particular adhesively bonded structures) is promising.

1) The interface wave produces shear stresses at the interface; these stresses are most sensitive to variations of adhesion quality.

2) The interface wave propagates along the interface and hence is sensitive to small changes in the properties of the adhesive and of the bond between the adhesive and the adherends.

3) The interface wave can be used for evaluation of very thin layers, when $2h/\lambda_t < 0.01$, where $2h$ is the thickness of the interface layer and $\lambda_t$ is the length of the shear wave in the substrate. Conversely, bulk longitudinal waves, usually employed for evaluation of adhesively bonded structures, are insensitive to the existence at the interface of a infinitely thin liquid layer which exhibits no shear resistance.

4) As shown below, interface waves can be used for evaluation of multilayered interface films, which simulate the adhesion properties at the adhesive-adherend interface.

As an example, reconstruction of the complex shear modulus of adhesive from experimental data is shown in figure 3. The data were taken in the course of bonding two steel substrates with epoxy resin. The thickness of adhesive film is 12μ.

![Fig. 3 The normalized complex shear modulus $\mu_0/\mu$ calculated from smoothed measured values of the interface wave velocity and the transmission losses. The dashed horizontal line represents direct measurements of the normalized film shear modulus $\mu_0/\mu$ after full polymerization. The steel shear modulus was taken as $\mu = 0.809 \cdot 10^{12}$ dyn/cm².](image-url)
THE CONCEPT OF THE EFFECTIVE SHEAR MODULUS - STRENGTH PREDICTION

An adhesive bond may fail in two ways: 1) Failure inside the adhesive, which is termed cohesive failure. The cohesive strength of a given adhesive correlates with its elastic modulus. 2) Failure along or close to the adhesive-adherend interface, which is termed interfacial or adhesion failure.

The cohesive strength of a given type of adhesive is governed by its chemical structure and by the conditions of the curing process (in particular, by the percentage of cross linking), and by the presence of various kinds of microdefects (e.g., microcracks or voids). These quantities also determine the shear modulus and the ultrasonic loss factor of the adhesive. By virtue of this fact, for a given type of adhesive, a change in the elastic modulus correlates with a change in its strength.

It is reasonable to consider a multilayered model of a bond line, consisting of an adhesive layer and thin interface layers between adhesive and adherends which characterize the adhesion properties.

According to this model, the shear modulus of the adhesive, calculated on the basis of the measured velocity and attenuation of interface waves is the effective shear modulus \( \mu_{\text{eff}} \). It characterizes the effective elastic properties of the multilayered adhesive system. The qualitative description of the properties of the interface wave can be given on the basis of the matrix method. We obtained a series expansion of the exact solution with respect to the thickness of interface films and compared this solution with the solution for the case of a single film (ref. 6).

According to this comparison, we obtained equation (1) for a single interface film when only linear terms are retained in the matrix expansion. A similar approximation for the multilayered system makes it possible to write the characteristic equation for the velocity of the interface wave in the form

\[
\frac{\mu_{\text{eff}}/\mu}{\Delta_\rho/(\beta - \frac{\rho_{\text{eff}}}{\rho} a_1 - 2\eta_2 a_2)}
\]

where

\[
\mu_{\text{eff}} = \mu_0 \left(1 + \frac{h_w}{h_0}\right) / \left(1 + \frac{h_w \mu_0}{h_0 \mu_w}\right); \quad \rho_{\text{eff}} = \rho_0 \left(1 + \frac{\rho_w h_w}{\rho_0 h_0}\right) / \left(1 + \frac{h_w}{h}\right)
\]

\( h = h_0 + h_w \)

2\( h_0 \) - is the thickness of the adhesive layer. \( h_w, \rho_w, \mu_w, \) - are the thicknesses, density and shear modulus of the weak boundary layer. The other terms have the same meanings as in Equation (1).

When contact between an adhesive and substrates is not ideal the interface wave velocity decreases. Hence, the effective shear modulus \( \mu_{\text{eff}} \) found from Equation (2) on the basis of the experimentally measured velocity will be smaller than the actual modulus of the film.

For example, in the case of absence of shear bonding between the adhesive and at least one of the substrates (slip contact) the velocity of the interface wave will be close to the Rayleigh wave velocity and the calculated \( \mu_{\text{eff}} \) will be equal to zero. It is thus seen that the interface-wave velocity will characterize not only the elas-
tic properties of the adhesive and its cohesive strength, but also the bond between the adhesive and the substrates, i.e., the interfacial (adhesion) strength. This means that the ratio $\mu_{\text{eff}}/\mu_0$ can be used as a criterion of the bond strength. If the effective shear modulus $\mu_{\text{eff}}$ is measured for a given bond, and the shear modulus $\mu_0$ of the adhesive is measured on a reference specimen, then Equation (3) can be used for estimating (ref. 7) the properties of the interfacial layer between adhesive and substrates.

EVALUATION OF THE CURING OF STRUCTURAL ADHESIVES BY ULTRASONIC INTERFACE WAVES

The reliability of adhesive-bonded structures and composite materials depends on a large number of different factors. It is determined to a significant extent by the manufacturing process and the feasibility of monitoring it. In connection with this it becomes necessary to control the quality of viscoelastic properties of the adhesive in the course of curing over the required temperature and pressure ranges, and to perform real-time continuous monitoring over the entire range of variation of adhesive properties in the course of the bond's performance.

The method of ultrasonic interface waves appears to be promising in resolving this problem.

The curing of structural adhesives is performed in nonisothermal conditions. The standard cure cycle consists of a rising temperature at a constant rate for 40 min. from room temperature to 120°C (for FM - 73) and to 180°C (for FM - 300 K). Temperature changes modify the elastic moduli of the adhesive film and substrates (adherends). To measure the adhesive properties under nonisothermal conditions, it is necessary to eliminate the effect of temperature variations in the substrates and transducers. For this purpose a differential measuring arrangement, in the form of an acoustic bridge, was suggested and analyzed (ref. 4). A computerized ultrasonic system was further developed to measure the phase difference of the ultrasonic signals from the two arms of the bridge.

The experimental sample in the form of an ultrasonic bridge is shown schematically in Fig. 4. Locations of the heaters and thermocouples are shown in the figure. The temperature gradient in the course of measurement did not exceed 0.5°C. As shown in the figure, pressure may be applied to the specimen. Strictly perpendicular force transmission is ensured by means of spherical contact. The comb transducers were used for excitation and reception of surface waves.

Fig. 4 Schematic of the specimen for differential measurements.  

Fig. 5 The block diagram of the computerized automatic measuring measurement.
The measuring arrangement is shown schematically in Fig. 5. A gated oscillator excites the transmitting transducer. The received signals are fed from two receiving transducers through attenuators and home-made dual-channel amplifier to a specially designed dual-channel phase-lock trigger. This unit is used for automatic following of changes in the phase of the signal between the working and reference channels. The received reference and working signals are transformed into zero-crossing rf signals. When one of the zeroes coincides with the selecting window, there is produced a short trigger/signal: trigger start, which starts up the counter. Analogously a second pulse: trigger stop, is produced in the second channel, and it stops the count. The change in the time shift between both triggers corresponds to a change in the phase between the working and reference signals. The temperature data and data from the counter operating in the time-interval averaging mode through IEEE-488 are fed to a microcomputer.

To check the accuracy of the system, we performed an analog simulation of the measuring method. For this a reference and a working signal from a highly stable frequency synthesizer were fed to the phase-lock trigger. The synthesizer's clock signal was used as the reference. The resulting time noise was measured to the order of 30 psec, which corresponds to the synthesizer error. This means that the noise of the electronic system, namely, the phase-lock trigger and counter, does not exceed 30 ps.

Some examples of application of the method for study of the curing of structural adhesives are given below. The changes of the phase velocity of interface wave during curing of FM-73 structural adhesive are shown in Fig. 6 by black circles. The shear strength data obtained in different stages of curing process are also shown in this figure. It is seen that a rise in the velocity of the interface wave (rise in the shear modulus of the adhesive) corresponds precisely to the time interval of the bond-strength growth. Fig. 7 shows results of the measurements at different temperatures. In Fig. 8 these results are summarized in the form of an Arrhenius plot from which the activation energy of the curing reaction for the FM-73 adhesive was found to be equal to 9.3 Kcal/mole. The data for FM-300K structural adhesive are shown in Fig. 9. The cure temperatures for this adhesive are higher than for FM-73.

The effect of aging of adhesive prepregs on cure are illustrated in Fig. 10 and Fig. 11. The adhesive was aged at 40°C before curing. The number of days of curing (24 hours in a day) are shown in Fig. 10 on the right side of the experimental curves. The summary results are presented in Fig. 11. Moderate aging results in appearance of nucleation sites, with shortening of the initial incubation period. For a longer time of aging prepreg became more polymerized and not converted to the fluid in the course of curing and therefore bond formation is not effective.

The technique discussed above is applicable to evaluation of thick bonded adherends. It was further developed (ref. 9) for evaluation of thin bonded sheets using Lamb waves.
Fig. 6 Comparison of the ultrasonic data with the variation in relative bond strength in the course of the cure.

Fig. 7 Data for FM - 73 adhesive at different temperatures.

Fig. 8 An Arrhenius plot of the cure data.

Fig. 9 Data for FM - 300K adhesive at different temperatures.

Fig. 10 The change in the interface-wave velocity as a function of curing time for aged specimens.

Fig. 11 The change in time delay as a function of the aging time.
When the thicknesses of both bonded substrates are much greater than a wavelength the interface wave can propagate along the interface as was discussed in the previous section. When the thicknesses of the bonded substrates are comparable to a wavelength the problem of wave propagation becomes much more complicated: several modes with different speeds can propagate in such a structure (Lamb modes). The number of modes and their phase and group velocities depend on frequency, substrate thicknesses and boundary conditions (adhesion) between substrates.

It is possible to select Lamb modes sensitive to the changes of the bond properties. Let us for example consider the dispersion characteristic of Lamb wave for two different boundary conditions: slip and rigid. The slip boundary condition corresponds to liquid adhesive on the interface (initial stages of curing). During the curing the adhesive solidifies and the boundary condition between plates becomes closer to the rigid boundary condition. During this liquid to solid conversion the propagated mode is transformed with change of phase velocity from a mode which can propagate for the slip boundary condition to one which can propagate for the rigid boundary condition.

As an example in Fig. 12 the dispersion curves for Lamb waves in a structure with slip and rigid boundary conditions are shown (two plates of the same thickness are bounded). The mode $S_0(h)$ can propagate in the case of slip bond. It is transformed to the mode $A_1(2h)$ during adhesive curing. It is shown schematically for $K_T = 3.1$ ($K_T$ is the wave number for shear waves, $h$ is the half thickness) by arrow.

Ultrasonic experiments and shear strength tests are usually performed on the lap-shear specimen Fig. 13(a) (ultrasonic measurements can be also performed for the sample shown in Fig. 13(b)). The wave behavior for such samples is much more complicated than was discussed above.

The Lamb wave is excited on the plate outside the bonded area. When it reaches the bonded area it is converted to the Lamb waves permissible in the plate region with a new value of the parameter $K_T$. This transformation will be different for different contact on the bondline (slip or rigid). So it is possible that some mode which can easily be excited for the slip contact cannot be excited at all for the rigid contact. So this mode will vanish in the course of curing of the adhesive and it cannot be used for our purposes.

To clarify this matter the theoretical analysis were applied for estimation of mode conversion on the boundary between bonded and unbonded areas (ref. 8). The numerical values of the energy transmission coefficients as functions of the parameter $K_T h$ are plotted in Fig. 14 ($2h$ is thickness of the bonded system). The first and last lower symbols shows the number of the mode in the bonded area. The middle lower symbol shows the incident and transmitted mode. The upper symbols show the type of this mode (symmetric(s) or antisymmetric(s)). The same curves can be used for slip contact: 1) antisymmetric waves may not be excited in the interface region 2) in the interface region modes with the same wave number as the incident wave can propagate in both sheets.

For the incident $S_0$ mode the energy is transported in the bonded region mainly by the mode $S_0(2h)$ at $0 < K_T h < 2$ for both rigid and slip contacts. Additionally, for slip contact, energy is also transported by the $S_0(h)$ mode (the same mode as incident).
$S_0 (2h)$ and $S_0 (h)$ modes will have different phase velocities as can be seen from the
dispersion caves shown in Fig. 12. For $K_{th} 2$ the energy in the bonded region is trans-
ported by the mode $A_1$ and by the mode $S_1$ when the bond is rigid (not shown in Fig. 14).
When the bond is slip the energy is transported by the $S_0(h)$ and $S_1$ modes. So with
changes of the type of contact the energy will be carried by different modes.

Fig. 12 The dispersion caves for the guided waves in the bonded region.

Fig. 13 Schematic illustration of the method of measurements and two sample config-
urations used.

Fig. 14 Energy transmission coefficients for incident $S_0(h)$ mode with $S_0(2h)$ mode
propagates inside the bonded region.
Now suppose that the properties of the interface change continuously from slip to rigid contact. This occurs when a thin film of liquid adhesive is cured on the interface. In which way can we attempt to select the incident mode and working frequency (parameter $K_{th}$) so that the selected mode will be sensitive to the interface properties and in particular to monitor the state of cure? We summarize here the main directions of analysis. 1) If the mode with the same phase velocity transports energy in the bonded region for slip and rigid contacts one can assume that the velocity changes for different bond quality will be small (this is clear because the difference between the limited values (slip and rigid) is negligible. 2) The energy transformation coefficients for the modes which are the main carriers of energy for slip and rigid contacts must be close in value. 3) The group velocities of the above modes must not be too different. Otherwise, the transmitted ultrasonic impulse for slip contact will be degraded if the contact changes and another signal will appear as the contact becomes rigid. In this case, it is impossible to make phase velocity measurements.

EVALUATION OF ADHESIVE CURING USING LAMB WAVES

The measurements were performed at frequencies 0.5, 1, 1.5, and 2 MHz. The thickness of the adhesive film is typically 80-90μ. We will discuss here only a few examples of the experimental data.

The experimental data for the incident $S_0$ mode at frequency 1 MHz are shown in Fig. 15. The time delay changes of the ultrasonic signal during adhesive curing are plotted.

The $K_{th}$ in the bonded area is about 3.2. When the adhesive is liquid the $S_0(h)$ is the main energy transporter. So the shift from the point 1 to the point 2 (Fig. 12) corresponds to the phase velocity changes during the polymerization. The phase velocity decreases and the time delay correspondingly increases.

Data for the incident $S_0$ mode at frequency 1.5 MHz are shown in Fig. 16. For rigid bond in the bonded area the energy transport is by the $A_1$ (2h) mode (parameter $K_{th} = 4.7$), and for slip contact by the mode $S_0(h)$. For this parameter $K_{th}$, the phase velocity of these modes were close to one another (practically equal). Experiment shows that the velocity changes are very small. The same measurements were done on the sample shown in Fig. 13(b). The changes of the phase velocity and attenuation are very close to those shown in Fig. 15 and Fig. 16. This supports the supposition that these changes are basically due to changes of the adhesive properties and the effect of the edges of the bond does no play a significant role.

Inversely the results for frequency 0.5 MHz (not shown here) are very different for the sample configuration shown in Fig. 13(a) and Fig. 13(b). One can assume that at this frequency the velocity changes occur not due to property changes on the bonded line but due to edge effects (ends of the bonded junction) because this is the only difference between the specimens shown in Fig. 13(a) and Fig. 13(b) from a wave propagation point of view. This may lead to the very interesting conclusion that some of the modes are sensitive to the edge condition. This is a very important from the fracture mechanics point of view.

Summarizing we can say that by selecting different working frequencies and incident modes for experiment, the different aspects of the bonding process can be studied. Some of the modes are sensitive to the bond line properties; some of them are more sensitive to the conditions of the edges of the bond.
Fig. 15 The phase velocity and transmitted amplitude changes during adhesive cure for $S_0(h)$ incident mode at frequency 1 MHz.

Fig. 16 The phase velocity and transmitted amplitude changes during adhesive cure for incident $S_0(h)$ mode at 1.5 MHz.

CONCLUSION

In this paper some results on application of interface and Lamb waves for study of curing of thin adhesive layers have been summarized. In the case of thick substrates (thickness much more than the wave length) the interface waves can be used. In this case the experimental data can be inverted and the shear modulus of the adhesive film may be explicitly found based on the measured interface wave velocity. It is shown that interface waves can be used for the study of curing of structural adhesives as a function of different temperatures and other experimental conditions. The kinetics of curing was studied. In the case of thin substrates the wave phenomena are much more complicated. It is shown that for successful measurements proper selection of experimental conditions is very important. This can be done based on theoretical estimations. For correctly selected experimental conditions the Lamb waves may be a sensitive probe of adhesive bond quality and may be used for cure monitoring.
REFERENCES