ULTRASONIC SPECTROSCOPY OF A LAYER
BETWEEN DISSIMILAR SUBSTRATES

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INTRODUCTION

Ultrasonic spectroscopy of a layer between two materials has been developed mostly for nondestructive testing of adhesive joints. For a thin layer separating two substrates signals reflected from front and back surfaces of the layer are overlapped in the time domain and interfere. The velocity of the ultrasonic wave and the attenuation in the layer can be obtained from analysis of the interference signal. Chang [1] and Flynn [2] used ultrasonic velocity and attenuation extracted from the ultrasonic reflected signal for correlation with the joint cohesive strength. The influence of different attenuation functions on amplitude and phase spectra of the signal reflected from the joint bondline was studied in [3]. Through-thickness resonance measurements were used in [4] to calculate both thickness and modulus of adhesive layer. An ultrasonic technique for evaluation of thin layers [5] and adhesive joints [6] was proposed by Kinra et al. A review of the field is given in [7]. In all the studies joints of similar substrates, predominantly Al/Al or steel/steel, were given the most consideration since these are widely used in industry. Emphasis was given to joint cohesive strength testing by normally incident ultrasonic waves. For adhesive/adherend interface evaluation ultrasonic spectroscopy at oblique incidence was developed [8, 9, 10]. Joints of similar substrates (Al/Al) were used for experiments; the data obtained was correlated with the joint adhesive strength. For general theory on elastic wave propagation in attenuating layered media the reader is referred to [11].

The work we present here addresses spectroscopy of adhesive joints of dissimilar materials. Interference of the ultrasonic signals reflected from the front and back of the adhesive layer results in appearance of minima in the spectrum of the signal reflected from the joint bondline. As one would expect from previous experience the increase of adhesive layer attenuation leads to widening and depth decrease of the spectrum minimum. We show that this is not always true for a layer between dissimilar materials. In certain conditions attenuation increase results in deepening and narrowing of the reflection minimum. In the following sections this phenomenon is studied theoretically and experimentally.
SPECTRA OF A LAYER BETWEEN DISSIMILAR SUBSTRATES

Anomalous Reflectivity of Attenuating Layer

Let us consider an ultrasonic plane wave normally reflected from a layer separating two similar substrates. This can be, for example, an adhesive layer in an adhesive joint of two aluminum plates. For calculation the thickness and properties of the adhesive layer are taken to be close to those met in practice: $h_{adh} = 135.8 \mu m$, $Z_{adh} = 2.5 \cdot 10^6 g/(cm^2 \text{sec})$. If the center frequency of the ultrasonic pulse is 5MHz signals reflected from front and back sides of the layer are not separated in time and interfere. Aluminum plates are thick enough so the signals reflected from the adherend surfaces not in contact with adhesive layer are separated in time from the interference signal. Minima in the spectrum of the signal reflected from the adhesive layer are due to destructive interference of the signals reflected from the front and back sides of the layer. Figure 1 shows calculated spectra for different attenuations in adhesive layer. At zero attenuation (i.e. when $\alpha = k''/k' = 0$, where $k'$ and $k''$ are real and imaginary parts of the wave number $k$) reflectivity at resonance equals zero. Increase of attenuation results in non-zero reflectivity at anti-resonance. Attenuation can be uniquely defined from the depth and from the width of the spectral minimum.

Figures 3(a,b) show calculated spectra for an ultrasonic wave reflected from the adhesive layer between aluminum and steel substrates (incidence from Al and steel sides). The thickness and properties of the adhesive layer are the same as in the previous case. At zero attenuation spectra obtained from aluminum and steel sides of the joint are identical. Reflectivities at anti-resonance equal that at the aluminum/steel interface: at resonance the adhesive layer is transparent to ultrasonic waves. Increase of attenuation leads to increase of reflectivity measured from the steel side, similar to the case of an Al/Al adhesive joint. It is opposite from the aluminum side: increase of attenuation initially leads to decrease of reflectivity (deepening of the reflection minimum). At “optimum” attenuation ($\alpha \approx 0.029$) reflectivity vanishes; further increase of attenuation results in reflectivity increase.

Figure 2 shows a schematic of ultrasonic wave reflection from an adhesive layer enclosed between two substrates. The incident signal has unit amplitude. Total reflection from the layer consists of two interfering signals: a) reflection from the top surface of the layer (first-reflection signal) with amplitude $R_{12}$ and b) reflection from the bot-

*indicates the complex value
When the upper substrate impedance is higher than that of the lower substrate and the attenuation in the layer equals zero the multiple-reflection signal amplitude is smaller than that of the first-reflection, thus at the anti-resonance the total reflectivity of the layer is not zero (Fig. 3(b), $\alpha = 0$). Increase of attenuation in the layer further decreases the multiple-reflection signal amplitude; thus reflectivity of the layer increases with attenuation.

When the upper substrate impedance is smaller than that of the lower and attenuation in the layer is zero the amplitude of the first-reflection signal is smaller than that of the multiple-reflection signal. At the anti-resonance increase of attenuation decreases the amplitude of the multiple-reflection signal (the first-reflection signal is almost unaffected) thus decreasing the amplitude of the reflected interference signal. At optimum attenuation the amplitudes of the first- and multiple-reflection signals are equal and the total reflectivity equals zero (Fig. 3(a)). Further increase of attenuation leads to further decrease of the multiple-reflection signal amplitude with consequent increase of the total layer reflectivity. It is interesting to note that similar effects were observed in optics for Fabry-Perot structures [12, 13] with asymmetric mirrors and absorption in the resonator cavity.

THEORY

Reflection from a Layer between Dissimilar Materials

Total reflection from a layer enclosed between two (in general different) materials
is given by

$$R^* = \frac{R_{12}^* - R_{32}^* e^{2ik'h}}{1 - R_{12}^* R_{32}^* e^{2ik'h}},$$

(1)

where $R_{ij}$ are complex valued stress reflection coefficients. At zero attenuation ($\alpha = k''/k' = 0$) all values in equation (1) are real. The reflection is a function of $k'h$ only; it is minimum at $k'h = \pi$ where it equals $R_{13}$.

At non-zero attenuation the reflectivity from the layer is a function of a complex wave number $k^*$ and can be represented as a function of two parameters: $k'h$ and $\alpha$. The anti-resonance condition ($k'h$ giving minimum reflectivity for a given attenuation) and the optimum attenuation (at which reflectivity at the anti-resonance equals zero) depend on impedances of the components of the layered structure.

Exact and Approximate Solutions

Complex valued reflection coefficients can be represented in the exponential form $R_{ij}^* = C_j e^{i\phi_j}$, $j = 1,3$, where and $C_j$, $\phi_j$ are functions of $\alpha$ and $R_{ij}$ and $T_{ij}$ (real stress reflection and transmission coefficients). For any given attenuation $\alpha$ there always exists $k'h$ such that the first- and multiple-reflection signals have opposite phases (anti-resonance condition):

$$k'h = \pi n + \frac{1}{2}(\phi_1 - \phi_3), \quad n = 1, 2, ....$$

(2)

$n$ is the order of the anti-resonance. In addition, for any given anti-resonance order $n$ there exists optimum attenuation $\alpha$ at which total reflection vanishes:

$$\alpha = \frac{1}{2k'h} \ln \left( \frac{C_3}{C_1} \right).$$

(3)

Equations (2) and (3) can be solved numerically to find for any given $n$ the phase $k'h$ and the attenuation $\alpha$ at which total reflection is zero. Representing these in polynomial form and keeping only terms not depending on $\alpha$ (usually $\alpha$ is less than 0.1) we obtain for the anti-resonance and optimum attenuation in the zero-order approximation

$$k'h \approx \pi n, \quad \alpha \approx \frac{1}{2\pi n} \ln \left( \frac{R_{32}}{R_{12}} \right), \quad n = 1, 2, ....$$

(4)
Equation (4) for $\alpha$ is valid only when $|R_{32}| > |R_{12}|$ or $Z_3 > Z_1$ (since $\alpha > 0$). The optimum attenuation is greatest for the first spectral minimum and in the limit $n \to \infty$ the optimum attenuation $\to 0$. When the impedance of the upper substrate is greater than that of the lower substrate zero reflectivity is impossible. In the first-order approximation the anti-resonance condition and the optimum attenuation are given by

$$k'h = \pi n + \frac{\alpha}{4} \xi, \quad \alpha = \frac{2\pi n}{\xi} \left[ \sqrt{1 + \frac{\xi}{2\pi^2 n^2} \ln \left( \frac{R_{32}}{R_{12}} \right)} - 1 \right], \quad n = 1, 2, \ldots \tag{5}$$

where $\xi = \left( \frac{1}{R_{32}} - R_{32} \right) - \left( \frac{1}{R_{12}} - R_{12} \right)$.

**Reflectivity from a Layer and Spectral Minimum Width**

Figure 4 shows the dependence of the total reflection at the anti-resonance from the polymer layer in an aluminum/polymer/steel sandwich on the layer attenuation for the ultrasonic wave incident from aluminum (solid line) and steel (dashed line) sides. From the steel side the reflection coefficient gradually increases with attenuation; it is much higher than that from the aluminum side and its dependence on attenuation is marked. From the practical point of view the low sensitivity of the reflection from the steel side to the attenuation means that the precision of the reconstructed attenuation will be low. For measurement from the aluminum side the dependence of the reflectivity on the layer attenuation is much greater with $R = 0$ at optimum attenuation. This means that the precision of attenuation measurement is considerably higher from the aluminum side but there will be two attenuation values satisfying the same reflectivity.

Additional information is needed to determine the attenuation of the polymer layer (when the measurement is done from the aluminum side). Figure 5 shows the dependence of the 3 dB width of the spectral minimum on the attenuation of the polymer layer. The minimum width equals zero at optimal attenuation. Dashed lines on the graph connect points corresponding to the same reflectivity (the same minimum depth); for the same reflectivity higher attenuation corresponds to a greater minimum width. The difference in corresponding widths is significant for large difference between attenuations and it is negligible when both attenuations are near the optimum value (and, consequently, to each other). Thus the minimum width can help determine the attenuation of the polymer layer if the two attenuations giving the same reflectivity are far from each other. When the two attenuations are close to each other phase spectra can be used to determine the correct value (an example will be shown in the Experimental section).

**Energy Loss**

The most physical measure of attenuation is energy loss; the higher the attenuation, the greater the loss of energy in the polymer layer. For an ultrasonic wave incident from material 1 and from material 3 the energy loss is given by

$$E_i = 1 - |R_i|^2 - |T_{i-3}| \cdot |T_{3-i}|, \quad \tag{6}$$

where $R_i$, $i = 1, 3$ are the total reflectivities from the polymer layer for incidence from side $i$ and $T_{i-j}$ are transmission coefficients through the layer for incidences from side 1 and side 3. Most of the energy stored in the resonator (represented by the polymer layer) is in the form of an ultrasonic wave at the anti-resonance frequency; thus the greatest energy loss occurs near the anti-resonance.

**EXPERIMENT**

Experiments were performed at normal incidence on different types of joints using ultrasonic goniometer described in [14]. Two types of polymer layers were used: 1) FM-73 epoxy resin prepreg manufactured by American Cyanamid, and 2) lexan polymer film. FM-73 prepreg film was used to prepare Al/Al and Al/Steel adhesive joint.
Adherend surface pretreatment and curing were performed according to aircraft specifications with resulting rigid adhesive/adherend interfaces and adhesive layer thickness $h \approx 110 \mu m$. Lexan polymer film was pressed between polished aluminum and steel plates with glycerin as couplant (with a resulting slip boundary at the metal/polymer interface). The outer edge of the resulting sandwich was sealed. A longitudinal wave at normal incidence interacts with the slip interface in the same way as with the rigid interface in the adhesive joint.

**Amplitude and Phase Spectra of a Reflected Ultrasonic Signal**

As an example of the experiments performed we describe here the results obtained for AI/steel joints. In the following discussion we refer to the AI/FM-73 adhesive/steel joint as Sample #1 and to the AI/lexan film/steel sandwich as Sample #2.

Figure 6(a) shows amplitude spectra of a longitudinal ultrasonic wave reflected normally from the adhesive layer in Sample #1. Circles correspond to measurement from Al side, squares to measurement from steel side. While the positions of the spectral minima are the same the depth observed from the steel side is much smaller than that from the aluminum side. As already discussed the depth of the minimum measured from the steel side uniquely defines the attenuation, but since the minimum depth measured from the steel side is only weakly dependent on attenuation (Fig. 4) the precision of the attenuation determined is low. The dependence of the minimum depth measured from aluminum on the polymer layer attenuation is much greater and can yields precise values of the attenuation. The difficulty is that two different attenuation values give the same minimum depth.

Figure 6(a) also shows spectra for incidence from the AI side calculated using two attenuations of the adhesive layer, giving experimentally measured reflectivity at the anti-resonance. In this particular case the two possible attenuations are very different ($\alpha_1 = 0.006$ and $\alpha_2 = 0.0743$), which results in significantly different minima widths (Fig. 5). Since the solid line in Figure 6 fits the experimental data much better than the dashed line, $\alpha = 0.0743$ is the attenuation of the adhesive layer. Figure 6(b) shows corresponding phase spectra. Again the solid line (corresponding to $\alpha = 0.0743$) fits experimental data much better than the dashed line ($\alpha = 0.006$).

It is more difficult to discriminate between two attenuations giving the same spectral minimum depth if their values are close to the optimum value and, thus, to each
other. Figure 7(a) shows spectra (experimental and calculated) of a normally incident longitudinal wave reflected from the aluminum side of the polymer film in Sample #2. Calculated spectra correspond to attenuations of the polymer layer of 0.0246 (dashed line) and 0.0381 (solid line). Since the minima widths for the calculated spectra are almost the same one cannot determine the attenuation corresponding to the polymer film. The determination can be done using phase spectra (experimental and calculated) shown in Figure 7(b). The attenuation of the polymer film should be $\alpha = 0.0381$ since the phase spectrum calculated using this value (solid line) fits the experimental data better.

Double Transmission and Determination of the Energy Loss in the Layer

To obtain a double transmission signal we acquired the signal transmitted through the sample reflected from the back-reflector and transmitted back to the transducer through the sample. The spectrum of the signal was deconvolved with that of the signal reflected from the back reflector and the frequency-independent losses at the sample/water and back-reflector/water interfaces were subtracted. Figure 8 shows the reflection and double-transmission spectra measured in the Al/steel adhesive joint (Sample #1). The calculated reflection and double transmission spectra corresponding to the two attenuations giving the same depth of the minimum for the reflectivity are shown in the graph by solid and dashed lines. The solid line corresponding to the actual adhesive layer attenuation fits well both the reflection and double transmission spectra. The dashed line evidently does not fit the experimentally measured double transmission spectrum. Although the double transmission spectrum defines the attenuation uniquely the precision of the experimental measurement is much lower than that for the measurement of the spectrum of the signal reflected from the adhesive layer due to additional losses at the water/sample and water/back reflector interfaces and the resulting low signal-to-noise ratio.

Figure 9 shows the frequency dependence of the energy loss calculated from the experimentally measured spectra of the reflected and double transmitted signals as well as theoretical curves for the two attenuations giving the same minimum depth in the reflectivity spectrum.

CONCLUSIONS

In this paper, the ultrasonic spectroscopy of a layer between two dissimilar materials is studied theoretically and experimentally. It is known that increase of the layer...
attenuation results in widening and depth decrease in the spectrum minimum of the reflected signal. This is not always true for a layer between dissimilar materials. It is shown that under certain conditions attenuation increase results in deepening and narrowing of the reflection minimum and that the layer reflectivity measured from the side of the substrate with lower impedance becomes zero at a certain “optimum” attenuation. Exact and approximate equations for the optimum attenuation and the antiresonance position are derived. Analogous phenomena were observed for both normal and oblique incidence. Based on the theoretical predictions an experimental method for the measurement of the layer attenuation is developed. The method uses both amplitude and phase spectra of the ultrasonic signal reflected from the layer. The possibility of using signal double transmission for energy loss measurement is also studied.

REFERENCES