ULTRASONIC CHARACTERIZATION OF THIN PLATE BONDING

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INTRODUCTION

Ultrasonic spectroscopy has been widely used for characterization of thin layered structures. Most previous studies \([1]–[12]\) have dealt mainly with analysis of the ultrasonic reflection or transmission spectra and the effect of bonding interfaces. More recently the inverse problem for interphase layer property determination from ultrasonic measurements has been addressed. Kinra and co-authors \([6], [9]\) use normally incident ultrasonic waves for determination of the longitudinal properties of the layer. In this case the elastic modulus, thickness and density are coupled and cannot be simultaneously recovered. The method for simultaneous determination of layer thickness, density, longitudinal and shear moduli and wave attenuations has been described in \([13]\). A thin layer between two thick substrates (Fig. 1a) was considered. Experimental data at two angles (normal and one oblique) was used for reconstruction.

Bonding of thin plates and foils with thermoset (adhesives) or thermoplastic polymers is widely used in industry. A typical example is packaging of food or medical products. Often in this application the metal foils have thin thermoplastic coatings and are bonded together with formation of a thin polymer interphase. The properties of this bonding layer are important characteristics in controlling the process of manufacturing during package sealing. Determination of all six properties of a thin polymer layer on a thin metal foil (Fig. 1b) has been described in the accompanying paper \([18]\). The problem of thin bonding layer property determination in the foil structure (Fig. 1c) is addressed in this paper. We call the structure thin when the reflected signals from all layers overlap and their interference must be taken into account. The method proposed allows one to determine all the properties of the interphase layer (layer 2 in Fig. 1c) – density, thickness, longitudinal and shear moduli and attenuations – using ultrasonic measurements at only two angles: one normal and one oblique. We assume the bond between layers perfect, the foil properties known and the interphase layer isotropic.

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Figure 1. Schematic of (a) thin layer bonded to thick substrates (semi-infinite substrates), (b) thin coating on thin substrate, (c) thin layer bonded to thin substrates. Shaded layers are substrates. V is the velocity of the substrate, τ is the pulse length.

THEORY
Definition of Six Nondimensional Parameters

The ultrasonic wave reflection (transmission) from a multilayered structure can be calculated using a matrix algorithm [14] whose applications to the bonding problem were discussed in [15, 16, 13]. In the model (Fig. 1c), we assume that the properties of substrates 1 and 3 are known. The reflection and transmission coefficients depend on six dimensional parameters of the unknown bonding layer 2: elastic moduli \((\lambda + 2\mu, \mu)\), thickness \(h\), density \(\rho\), and longitudinal and shear wave attenuations \((\alpha_\ell, \alpha_t)\): 

\[
\lambda + 2\mu, \mu, \rho, h, \alpha_\ell, \alpha_t.
\]

To simplify the inversion of the parameters from ultrasonic data, we introduce six nondimensional parameters:

\[
Z_N, \bar{h}_\ell, \bar{h}_{\theta\ell}, \bar{h}_{\theta t}, \alpha_\ell, \alpha_t,
\]

They are defined as normalized impedance \(Z_N = Z_2/Z_1\), nondimensional thickness at normal incidence \(\bar{h}_\ell = \omega_0 h/V_\ell\), nondimensional thicknesses at oblique incidence \(\bar{h}_{\theta\ell} = \omega_0 h \cos \theta_\ell/V_\ell\) and \(\bar{h}_{\theta t} = \omega_0 h \cos \theta_t/V_\ell\), and longitudinal and shear wave attenuations \(\alpha_\ell = k_\ell''/k_\ell\) and \(\alpha_t = k_t''/k_t\), where \(Z_1, Z_2\) are impedances of the substrate 1 and the unknown layer 2, \(V_\ell = [(\lambda + 2\mu)/\rho]^{1/2}\), \(V_t = [\mu/\rho]^{1/2}\) are longitudinal and shear wave velocities in the unknown layer, \(\theta_\ell, \theta_t\) are corresponding propagation angles, and \(\omega_0 = 1\) MHz.

All six nondimensional parameters (2) can be found from two measurements: one at normal and the other at oblique incidence by the inversion algorithm described in the next section. Four nondimensional parameters from the set (2) \((Z_N, \bar{h}_\ell, \bar{h}_{\theta\ell}, \bar{h}_{\theta t})\) fully define four dimensional parameters from the set (1):

\[
\begin{align*}
\lambda + 2\mu &= \frac{Z_N Z_1 \sqrt{\bar{h}_\ell^2 - \bar{h}_{\theta\ell}^2}}{\xi_0 \bar{h}_\ell}, \\
\mu &= \frac{Z_N Z_1 \bar{h}_\ell \sqrt{\bar{h}_\ell^2 - \bar{h}_{\theta\ell}^2}}{\xi_0 \bar{h}_\ell^2 - \bar{h}_{\theta\ell}^2 + \bar{h}_{\theta t}^2}, \\
\rho &= Z_N Z_1 \xi_0 \frac{\bar{h}_\ell}{\sqrt{\bar{h}_\ell^2 - \bar{h}_{\theta t}^2}}, \\
h &= \frac{\sqrt{\bar{h}_\ell^2 - \bar{h}_{\theta t}^2}}{\xi_0 \omega_0},
\end{align*}
\]

where \(\xi_0 = \sin \theta_0/V_0\) and \(\theta_0, V_0\) are propagation angle and wave velocity in water. Instead of the elastic moduli, the longitudinal and shear wave velocities, \(V_\ell\) and \(V_t\), can be used. They
are related to the nondimensional parameters by

{\begin{align*}
V_t & = \frac{\sqrt{h_t^2 - h_{t0}^2}}{\xi_0 h_t}, & V_i & = \frac{\sqrt{h_i^2 - h_{i0}^2}}{\xi_0 \sqrt{h_t^2 - h_{t0}^2} + h_{o0}^2}.
\end{align*}}

(5)

Inversion algorithm

As discussed in [13], the reflection spectrum at normal incidence is fully defined by three nondimensional parameters: \( Z_N, \overline{h}_t \) and attenuation \( \alpha_L \), which can be determined from experimental data by inversion. \( Z_N \) and \( \overline{h}_t \) are functions of three dimensional parameters: \( \lambda + 2\mu, h \) and \( \rho \). Two of them can be determined only if the third is known: this is the main limitation of using only normal incidence measurements for layer characterization. To determine all layer properties (1) oblique incidence measurements are also required. It was proposed in [13] to use measurements of both normal and oblique incidence reflection (transmission) spectra for determining six layer properties (1) with no prior knowledge. For the nondimensional set of parameters (2) the problem is factorized (decomposed) and a two-step algorithm for determination of the adhesive layer properties in a three-dimensional space of parameters can be used. First, we determine three nondimensional parameters: \( Z_N, \overline{h}_t \) and \( \alpha_L \), from reflection (transmission) spectra at normal incidence. Next, considering \( Z_N, \overline{h}_t \) and \( \alpha_L \) as known, three more nondimensional parameters \( \overline{h}_{t0}, \overline{h}_{o0}, \alpha_t \) are determined from oblique incidence data (reflection or transmission). The corresponding dimensional parameters (1) are calculated using equations (3)-(4).

For inversion we employ the least squares method for the minimization of the sum of squared deviations between experimental and calculated reflection (transmission) coefficients considering nondimensional parameters (2) as variables in a multidimensional space:

{\begin{align*}
\min_{X_i \in \mathbb{R}^n} \frac{1}{2} \sum_{i=1}^{m} (|R_i^e| - |R_i^c|)^2
\end{align*}}

(6)

Here \( X_i \) are nondimensional parameters (2), \( n = 3 \) is the number of parameters to be found, \( m \) is the number of data points at different frequencies, and \( R^e \) and \( R^c \) are the experimental and calculated reflection (transmission) coefficients, respectively.

SENSITIVITY OF THE REFLECTION COEFFICIENT TO THE LAYER PROPERTIES

In general, the inverse problem is well-posed if its solution (a) exists, (b) is unique, and (c) is stable, i.e., to small changes of experimental data (input) correspond small changes of the solution (output). If the solution does not satisfy these conditions, the problem is called ill-posed and its treatment requires special methods. The main focus of the further discussion is a) to find the frequency range in which the ultrasonic reflection (transmission) signal is most sensitive to the interphase layer properties and b) to demonstrate that in that frequency range the layer properties can be found using the proposed algorithm from the measured spectra at normal incidence and one oblique incidence angle. We take a pragmatic approach. We consider the inverse solution unique if the least-squares minimization converges, whatever the initial guess, to the same solution. We consider the solution stable if it is weakly dependent on random scatter of the experimental data. Another stability consideration is reflection (transmission) coefficient sensitivity to a parameter \( p \) of the layer. Let us assume explicit dependence of the reflection (transmission) coefficient on the layer parameter \( p, R = \overline{R}(p) \). Then the relative errors in \( p \) and \( R, \epsilon_R = \delta |R|/|R|, \epsilon_p = \delta p/p \), are related by

{\begin{align*}
\epsilon_p & = \epsilon_R/X_{R,p},
\end{align*}}

(7)

where \( X_{R,p} \) is the sensitivity of \( R \) to \( p \); it is defined as:

{\begin{align*}
X_{R,p} & = \frac{p}{|R|} \frac{\partial |R|}{\partial p}.
\end{align*}}

(8)

If \( X_{R,p} \ll 1 \), then amplification of error occurs. Thus, the sensitivity parameter is a good indicator of inversion stability. This is in line with the intuitive notion that if the reflection
Figure 2. a) Normal ultrasonic reflection spectra from the Al(70 μm)/Polypropylene(54 μm)/Al(110 μm) laminate and b) corresponding sensitivity curves for $Z_N$, $h_f$ and $\alpha_f$. The properties of the polypropylene layer are: $(\lambda + 2\mu) = 5.45$ GPa, $\mu = 1.23$ GPa, $\rho = 0.99$ g/cm$^3$, $h = 0.054$ mm, $V_f = 2.35$ km/sec, $V_t = 1.11$ km/sec, $\alpha_f = 0.023$, $\alpha_t = 0.600$. Wave incident from the 70 μm Al foil side.

Figure 3. a) 20° ultrasonic oblique reflection spectra from Al(70 μm)/Polypropylene(54 μm)/Al(110 μm) laminate and b) corresponding sensitivity curves for $h_{bf}$, $h_{bt}$, and $\alpha_f$. Polypropylene properties are the same as in Fig. 2. Wave incident from the 110 μm Al foil side.

(transmission) coefficient is weakly dependent on one of the layer parameters, this parameter is difficult to find from the reflection (transmission) coefficient measurement.

Selection of the Frequency Range

For efficient determination of the bonding layer properties, it is important to perform measurements in the frequency range with higher sensitivity of the reflection coefficient to the layer properties. Fig. 2a shows a calculated spectrum of the reflection coefficient at normal incidence; Fig. 2b shows corresponding sensitivity curves for the nondimensional parameters $Z_N$, $h$ and $\alpha_f$. The first reflection minimum (Fig. 2a) at about 5 MHz is deeper than the minima at 21 and 30 MHz and therefore has the highest signal to noise ratio. From the sensitivity curves in Fig. 2b, one finds that the first reflection minimum also has the highest sensitivity for $Z_N$, $h$ and $\alpha_f$. Therefore the frequency range from 3 to 8 MHz is selected for normal incidence measurements. The calculated reflection spectrum at 20° oblique incidence and corresponding sensitivity curves are shown in Fig. 3a and 3b. The frequency range from 13 to 23 MHz is selected as the oblique incident measurement frequency range.
Figure 4. a) Spectra of ultrasonic signal reflected from the Al/PP/Al laminate at normal incidence and b) change of the depth of minima at different longitudinal attenuation $\alpha_i$.

Figure 5. Spectra of ultrasonic signal reflected from the Al(73 $\mu$m)/PP(46 $\mu$m)/Al(118 $\mu$m) laminate at 20° oblique incidence.

EFFECT OF THE SIDE OF ULTRASONIC WAVE INCIDENCE

It was found that for the laminate with top and bottom substrates of different thickness, the reflection minimum depth depends on the sample side irradiated by the incident beam. This is similar to the effect described in [17] for the layer bonded by two different semi-infinite substrates. As an example, the experimental results and calculation for a Al(73 $\mu$m)/PP(46 $\mu$m)/Al(118 $\mu$m) laminate system are shown in Fig. 4a for normal incidence. The first resonance is much deeper for the case of beam incidence from the thinner foil side than from the thicker foil side. The reflection minimum is caused by destructive interference of the signals reflected from each layer. The effect of the PP layer attenuation on the minimum depth at first resonance for incidence from the different sides of the sample with different foil thickness and for the sample with identical top and bottom foils is shown in Fig. 4b. The layer attenuation for deepest minimum is called the critical attenuation [17]. For a laminate with identical aluminum foil thickness, the minimum depth decreases monotonically when attenuation increases. To improve signal to noise ratio for normal incidence measurements, we select the wave to be incident from the thin foil side.

Fig. 5 gives reflection spectra for an ultrasonic beam incident at 20° from different sides of the laminate. In the frequency range 13 to 23 MHz, the reflection minima are much more
pronounced for the case of beam incidence from the thicker foil side than from the thinner foil side. The reverse effect occurs at higher frequencies (25 to 37 MHz) where deeper minima occur for incidence from the thinner foil side. However, in this frequency range lower signal to noise ratio is observed in our experimental conditions and for the oblique incidence case we select the measurement frequency range 13 to 23 MHz with the beam incident from the thicker foil side.

**STABILITY OF INVERSION ALGORITHM TO SCATTER IN EXPERIMENTAL DATA**

To analyze the possible errors in the reconstruction of the bonding layer parameters, we studied the stability of the inversion algorithm. A set of polymer film properties ("original set") is used to generate numerically synthetic reflection spectra at normal and oblique incidence. The spectra are overlapped with a typical transducer spectrum. A backward FFT procedure is used to calculate the corresponding synthetic time-domain signal. Next, different levels of random noise are introduced into the time-domain signals to simulate possible experimental noise. A forward FFT procedure is applied to the "noisy" time-domain signals and deconvolved with the transducer spectrum to obtain synthetic "noisy" reflection spectra. These spectra are used to determine the elastic constants by the nonlinear least-square optimization method discussed previously. The spectral data from the frequency ranges determined by the sensitivity analysis are used for reconstruction.

Due to the introduced noise the reconstructed set of polymer film properties is not exactly equal to the original set. It is compared to the original set to study the effects of the noise level. For each noise level the procedure is repeated 400 times, the reconstructed parameters (dimensional and nondimensional) are normalized to the original value and the average $\bar{\rho}$ and standard deviation $\Delta \rho$ are calculated for the normalized values.

The spectral data from the frequency ranges determined by the sensitivity analysis are used to investigate the stability of the inversion algorithm to the scatter in the experimental data. Table 1 summarizes the reconstructed non-dimensional and dimensional parameters for 1% noise level. It compares the reconstructed results of thin substrate (laminate) with the results for a layer between semispaces [13]. One sees that at the same noise level of 1%, the errors of the parameters determined for the polymer film between the thin foils are larger.

<table>
<thead>
<tr>
<th>Sample</th>
<th>laminate</th>
<th>semispace substrates</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Z_N/Z_N^0 (\epsilon_{Z_N})$</td>
<td>0.997 (2.3%)</td>
<td>1.000 (0.4%)</td>
</tr>
<tr>
<td>$\epsilon_l/\epsilon_l^0 (\epsilon_{l_l})$</td>
<td>0.997 (2.1%)</td>
<td>1.000 (0.0%)</td>
</tr>
<tr>
<td>$\epsilon_{gel}/\epsilon_{gel}^0 (\epsilon_{gel})$</td>
<td>0.995 (4.2%)</td>
<td>1.000 (0.1%)</td>
</tr>
<tr>
<td>$\epsilon_{gel}/\epsilon_{gel}^0 (\epsilon_{gel})$</td>
<td>1.001 (0.8%)</td>
<td>1.000 (0.1%)</td>
</tr>
<tr>
<td>$\mu/\mu^0 (\epsilon_{\mu})$</td>
<td>0.994 (3.4%)</td>
<td>1.000 (0.4%)</td>
</tr>
<tr>
<td>$\rho/\rho^0 (\epsilon_{\rho})$</td>
<td>0.996 (7.6%)</td>
<td>1.001 (0.7%)</td>
</tr>
<tr>
<td>$h/h^0 (\epsilon_{h})$</td>
<td>1.001 (3.0%)</td>
<td>1.000 (0.5%)</td>
</tr>
<tr>
<td>$V_l/V_l^0 (\epsilon_{V_l})$</td>
<td>1.005 (5.2%)</td>
<td>1.000 (0.5%)</td>
</tr>
<tr>
<td>$V_l/V_l^0 (\epsilon_{V_l})$</td>
<td>1.004 (2.1%)</td>
<td>1.000 (0.4%)</td>
</tr>
<tr>
<td>$\alpha_l/\alpha_l^0 (\epsilon_{\alpha_l})$</td>
<td>1.000 (0.7%)</td>
<td>1.001 (1.0%)</td>
</tr>
<tr>
<td>$\alpha_l/\alpha_l^0 (\epsilon_{\alpha_l})$</td>
<td>1.005 (2.6%)</td>
<td>1.005 (1.3%)</td>
</tr>
</tbody>
</table>

Table 1. Average values and standard deviations of the normalized parameter reconstructed from simulated 'noisy' spectra with 1% noise levels. Angle of oblique incidence is 20° for thin foil laminate case and 17° for semispace substrates case. Data for the semispace substrates case come from reference [13]. Original values of the dimensional parameters for polypropylene are: $(\lambda + 2\mu)^0 = 5.45$ GPa, $\mu^0 = 1.23$ GPa, $\rho^0 = 0.99$ g/cm$^3$, $h^0 = 0.054$ mm, $V_l^0 = 2.35$ km/sec, $V_t^0 = 1.11$ km/sec, $\alpha_l^0 = 0.023$, $\alpha_l^0 = 0.060$. Thin Al foil thickness = 70 μm, thick Al foil thickness = 110 μm.
than those of the polymer between the semi-infinite substrates. This is because the multiple reflections in the thick substrate can be separated from the interface reflection, and only the effect of wave interference in the polymer layer is measured. On the other hand, for the laminate case, the wave interference in thin foil substrates and polymer layer is coupled and the polymer layer properties are reconstructed from the reflection spectrum of the whole laminate with less precision. Despite the larger errors for the laminate case, the error is within a few percent for the 1% noise level which is reasonable for many practical cases.

EXPERIMENT

Sample Preparation Procedure

A typical laminate structure is bonded by a heating and pressuring process using polypropylene as the bonding material (usually the polypropylene layer is coated on one of the aluminum foils). We simulated the heating and pressuring process in our sample preparation. Two kinds of aluminum foils (4x4 cm) were used in preparation. One is 110 µm Al foil coated with polypropylene layer. The other is 70 µm Al foil. Two aluminum foils were put together with the polypropylene layer between the two aluminum foils. They were heated to 220°C in 10 minutes under pressure 3MPa. Then the pressure was released and the laminate cooled down slowly to the room temperature. Two kinds of samples were prepared. Sample A has different substrate thickness, 70 µm Al foil/56 µm polypropylene/110 µm aluminum foil. Sample B has the same substrate thickness, 114 µm aluminum foil/67 µm polypropylene/114 µm aluminum foil.

Bonding Layer Properties Determination for the Laminate Samples

Fig. 6a and b are the experimental results for the prepared thin foil laminate sample A. The frequency range used for normal incidence measurement is 2.8 to 5.7 MHz. The frequency range used for oblique measurement is 13 to 23 MHz. Nondimensional parameters $Z_N$, $h$ and $\alpha_f$ are obtained from the normal measurement data. Then they are used as known parameters in determination of three other nondimensional parameters $\bar{h}_{qT}$, $\bar{h}_{qH}$, and $\alpha_f$ from oblique data. The six dimensional parameters then are calculated from these nondimensional parameters (Eq. (3)-(4)). Table 2 gives the reconstructed properties for both sample A and B. One sees that for samples prepared at the same conditions but with different substrate geometry (thickness) the reconstructed polypropylene properties agree with each other very well. The directly measured thickness of the polypropylene layers also agrees with the ultrasonic measurement results.
Table 2. Reconstructed properties of the polypropylene (PP) layer in the laminate sample A(70 μm Al/54 μm PP/110 μm Al) and sample B(114 μm Al/67 μm PP/114 μm Al). The thickness in brackets is measured directly by micrometer.

<table>
<thead>
<tr>
<th></th>
<th>λ + 2μ, GPa</th>
<th>μ, GPa</th>
<th>ρ, g/cm³</th>
<th>h, mm</th>
<th>αx</th>
<th>αt</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample A</td>
<td>5.45</td>
<td>1.23</td>
<td>0.99</td>
<td>0.054(0.056)</td>
<td>0.023</td>
<td>0.060</td>
</tr>
<tr>
<td>Sample B</td>
<td>5.42</td>
<td>1.23</td>
<td>1.03</td>
<td>0.067(0.070)</td>
<td>0.034</td>
<td>0.071</td>
</tr>
</tbody>
</table>

CONCLUSIONS

This paper describes experimental and theoretical study of an ultrasonic method for determination of the properties of a polymer layer inside a foil laminate (thickness less than the ultrasonic wavelength). The method allows simultaneous determination of all properties — thickness, density, longitudinal and shear elastic moduli and attenuations — from two measured reflection spectra: one at normal and one at oblique incidence. The sensitivity of the reflection spectrum to the individual properties and the stability of the inversion method against experimental noise are studied. Based on the sensitivity analysis, measurement frequency ranges were selected (3-8 MHz for normal incidence, and 13-23 MHz for oblique incidence). It is shown that the polymer properties can be reconstructed from the laminate reflection spectrum with less precision than those determined for the layer bonded by semi-infinite substrates. Despite the larger errors for the laminate case, the error is within a few percent for the 1% data scatter which is reasonable for many practical cases. It is found that the depth of the reflection spectrum minimum depends on the side of ultrasonic beam incidence and thus affects the precision of the properties determination. For the structure studied the measurements were performed from the thin Al foil side for normal incidence and from the thick Al foil side for oblique incidence. The application of the proposed ultrasonic method is demonstrated for experimental determination of the properties of a polypropylene layer between thin Al foils.

REFERENCES