NONLINEAR ANGLE BEAM ULTRASONIC EVALUATION OF ADHESIVE BONDS

S. I. Rokhlin¹, L. Wang¹, A. Baltazar¹, V. A. Yakovlev² and L. Adler²

¹The Ohio State University, 1248 Arthur E. Adams Dr, Columbus, OH 43221
²Adler Consultants, 1275 Kinnear Road Columbus, OH 43212

ABSTRACT. We have developed an experimental method incorporating high frequency pulsed angle beam ultrasonic measurements under low frequency vibration of bonded structures utilizing parametric/nonlinear mixing between high and low frequencies. We have demonstrated that the effect of environmental degradation of adhesive bonds can be detected by this method. It is shown that good quality (undamaged) bonds do not exhibit dependence of their ultrasonic signatures on the overlay of low frequency vibration loads; however, environmentally degraded or imperfect bonds exhibit a shift of the resonance frequency of the ultrasonic signal reflected from the bond. We have also found that a much higher level of mixing (two orders of magnitude) occurs for very brittle adhesive bonds whereas the linear method cannot discriminate these bonds. We have developed a model for the case of two nonlinear interfaces separated by an adhesive layer and have described the variations in the reflection coefficient associated with nonlinear interfacial spring density changes for normal and oblique incidence of longitudinal or transverse waves. We have simulated second harmonic generation as a function of the spring density and also developed a model and predicted parametric mixing of the high frequency reflected wave and the low frequency interface vibration excitation.

INTRODUCTION

Adhesive joining of materials has found widespread use in the aerospace industry[1]. However, lack of reliable nondestructive testing methods limits use of adhesive joining in critical structural applications. The integrity of an adhesive joint depends on the interfacial properties between the adhesive and adherents and on the bulk properties of the adhesive. Premature failure can be either cohesive or adhesive (interfacial) in nature depending on whether fracture occurs in the interior of the adhesive layer itself or very close to the adhesive/adherent interface. Weak bonds remain hidden from conventional inspection methods, which reliably detect only gross defects such as voids and open delaminations.

Several studies [2-5] describe ultrasonic nonlinear measurements of adhesive bonds. While the approach shows some promise, significant effort is still needed to transform it into a quantitative NDE tool. Environmental degradation of adhesive joints of composite structures affects the adhesive/adherent interface by decreasing the number of molecular bonds between the adhesive and the substrates. In this paper we will model the mechanical bond on the adhesive adherent interface by distributed spring boundary conditions. In this model, the spring density becomes a function of the applied stress,
leading to nonlinear behavior of the imperfect interface. Since the spring density is related to the molecular structure at the interface, this approach potentially leads to evaluation of the local bond strength. We will also describe a nonlinear ultrasonic method for quantitative evaluation of the spring bond density at the adhesive/substrate interface using dual beam ultrasonic spectroscopy under superposed dynamic load.

THEORY

Nonlinear Model of an Adhesive Bond

Nonlinear acoustic phenomena are often associated with defects or local disorder. In the case of interfaces, molecular level disbonds or imperfections at the boundary can lead to nonlinear interfacial behavior. During curing, the thermosetting adhesive forms a three dimensional lattice where large polymer chains are connected by epoxy groups. At the interface, these epoxy groups form hydrogen and Van der Waals bonds with the substrates. As a result of environmental degradation or poor surface condition, the molecular bond density can be significantly reduced locally leading to bond strength reduction and interfacial failure.

To characterize the molecular bond density change, we use a variable equivalent spring density, which is modeled by a set of identical springs of various lengths [6] as shown in Fig. 1. The applied stress varies the number of springs in contact. Thus the spring density becomes a function of the applied stress, leading to nonlinear behavior of the imperfect interface. The boundary conditions at this nonlinear interface may be written as

\[ \sigma^+ = \sigma^- = K(P, \Delta U) \Delta U, \]

where \( \sigma^+ \) and \( \sigma^- \) are stresses generated by the high frequency ultrasonic wave at the top and bottom surface of the interface, and are continuous across the interface. \( \Delta U \) is the displacement jump across the interface and \( P \) is the external low frequency modulation force. Because \( \Delta U \) is the displacement jump corresponding to the high frequency ultrasound and is small, we can expand the spring density \( K(P, \Delta U) \) as a series in \( \Delta U \).

Taking the first two terms, we have

\[ K(P, \Delta U) = K_0(P) \Delta U + \frac{1}{2} \Delta U^T \beta(P) \Delta U, \]
where $K_0(P)$ and $\beta(P)$ are the linear and nonlinear terms respectively. These parameters depend on interface properties and a function of the applied stress $P$: for a weak bond, nano-separations occur under tension in areas of reduced density of molecular bonds and new failures of the molecular bonds may occur due to increased load per bond. The threshold level of maximum stress at which this decrease occurs can also be used to characterize the number of molecular bonds. We assume that in a good bond with a normal density of molecular bonds, the molecular bonds will not change under stress leading to an unchanged spring density under applied stress. Thus the measured spring density can be related to the quality of the adhesive bond and the local microscopic strength.

Second Harmonic Generation for Ultrasonic Wave Interaction with Nonlinear Interfaces

First let us consider plane harmonic wave interaction with a nonlinear interface. The nonlinear boundary conditions are expressed as Eq. (1). The incident wave is a harmonic plane wave. Due to nonlinearity, the reflected and transmitted waves include all harmonics and may be written as the summation of all harmonic plane waves

$$U_R = u_1(\omega)e^{i(k_1x-\omega t)} + u_2(2\omega)e^{i(k_2x-2\omega t)} + \ldots,$$

$$U_T = u_1(\omega)e^{i(k_1x-\omega t)} + u_2(2\omega)e^{i(k_2x-2\omega t)} + \ldots.$$ (3)

Substituting all these equations into the boundary conditions (2), one can obtain the first and higher harmonic reflection-transmission coefficients. For normal incidence, the first harmonic reflection and transmission coefficients may be written as

$$R_{12}(\omega) = \frac{1}{Z_1} - \frac{1}{Z_2} + i\omega/K_u,$$

$$T_{12}(\omega) = 1 + R_{12}(\omega),$$ (4)

the second harmonic reflection coefficient is

$$R_{12}^{2}(2\omega) = \frac{\beta}{2\omega^2} \left( \frac{1}{Z_1^2} (1 - R_{12})^2 + \frac{1}{Z_2^2} T_{12}^2 - \frac{2}{Z_1 Z_2} (1 - R_{12}) T_{12} \right).$$ (5)

where $Z_i = \rho_i V_i$ is the acoustic impedance, $\rho_i$ is density and $V_i$ is sound velocity in medium $i$.

Ultrasonic wave interaction with an adhesive joint was analyzed similarly. In this case, the wave field inside the adhesive layer is also expressed as the summation of all harmonic up-going and down-going plane waves and the boundary conditions at the top and bottom surfaces of the adhesive layer are all represented by Eq. (1). Substituting the incident, reflected, transmitted waves and the waves inside the layer into both interface conditions and solving these equations for different harmonics, we obtain the reflection and transmission coefficients for different harmonics. Fig. 2 shows the reflection and transmission coefficients at normal incidence on the interface between two contacting aluminum substrates. Figure 2a presents the results for the first harmonics. As expected, the reflection coefficients decrease as load increases. The second harmonic reflection
FIGURE 2. Reflection and transmission coefficients for a nonlinear interface as function of pressure. The two substrates are aluminum. (a) first harmonics, (b) second harmonics.

FIGURE 3. Example of frequency dependent reflection coefficient for first harmonic (top), for incident second harmonic excited by ultrasonic transducer (middle) and second harmonic generated at the interface. One should note that the reflected second harmonic (middle figure), which may be generated due to nonlinearity of the transducer, has interference resonance dips while the interface-generated second harmonic (bottom) has peaks. This may help to decrease the effect of the second harmonics due to nonlinearity of the system while recording nonlinear response of the interface.

coefficient shown in figure 2b has a peak around 10MPa. Comparing those two figures one sees that the second reflection peak appears where the first harmonic reflection has the largest slope. Figure 3 shows the results for an adhesive bond (Aluminum + Adhesive + Aluminum). The thickness of the adhesive is 0.2mm. One can see the resonance appears as a dip in the first harmonic reflection. In the second harmonic reflection, it appears as a peak.

Mixing of High and Low Frequency Vibration

We also developed a model to describe the interaction between a high frequency ultrasonic wave and a nonlinear interface under low frequency oscillatory loading. To
describe this problem we derived the time-dependent boundary conditions for the reflection stress $A_R$:

$$\frac{dA_R}{dt} = -K_n(t)(A_I(1/Z_1 - 1/Z_2) + A_R(1/Z_1 + 1/Z_2)) + \frac{\beta}{2K_n(t)}(A_I + A_R)(A_I(1/Z_1 - 1/Z_2) + A_R(1/Z_1 + 1/Z_2)),$$

(6)

where $Z_1$ and $Z_2$ are the acoustic impedances of the upper and bottom semispaces. $A_I$ is the stress corresponding to the incident wave. The higher frequency wave reflected from the interface with the nonlinear time-dependent boundary condition will be mixed with an external modulation vibration of frequency. The nonlinear equation (6) can be solved numerically by the Runge-Kutta method in the time domain or analytically by the harmonic analysis method in the frequency domain.

**Angle Beam Reconstruction for Interfacial Stiffness**

Wang et al. [7] has developed an algorithm to determine both the interfacial springs and bulk adhesive layer properties using ultrasonic measurements [8-11]. In this algorithm, these properties are divided into two groups, one of them related to the normal incidence reflection spectra and other to the oblique spectra. Each group has four nondimensional parameters. First the normal and oblique spectra are measured experimentally. Second the first group of parameters is obtained from the normal spectrum using a least square optimization algorithm. Then the oblique reflection spectrum together with the first group reconstructed parameters are used to obtain the second group parameters. After all nondimensional parameters are obtained, the dimensional parameters describing the interfacial springs and adhesive layer bulk properties are determined.

**EXPERIMENT**

**Experimental Apparatus for Stress Modulated Angle Beam Ultrasonic Spectroscopy**

The schematic of the experimental setup for stress modulated angle beam ultrasonic spectroscopy is shown in Figure 4. The low frequency vibrations are exited by a piezoelectric high power ultrasonic transducer manufactured by Branson for ultrasonic welding. The resonance frequency of the unloaded transducer is about 20 kHz. The vibration signal is applied from a function generator to the shaker through a power amplifier and impedance matching unit. The bonded sample is attached rigidly to the shaker. The vibration amplitude on the sample is measured by an accelerometer. The bonded sample consists of two Al blocks as shown in Figure 4. The top aluminum block holds three transducers that allow measurements of reflection coefficients at normal and oblique incidence. This setup was used for measurements of the reflected ultrasonic signal mixing with low frequency vibrations. For perfect bonds no mixing is expected; for weak interfaces the boundary conditions change under load leading to mixing higher and low frequency vibrations. Both continuous and pulse high frequency ultrasonic signals were used in these experiments.
FIGURE 4. Schematic of the experimental setup for stress modulated ultrasonic spectroscopy in vibration mixing mode. Frequency of the low frequency vibration modulation is 15-20 kHz.

FIGURE 5. Simulated and experimental modulation spectra.

Results and Discussion

The simulated (using Eq. 6) and experimental spectra of the reflected ultrasonic signal under low frequency stress modulation are shown in Fig. 5. The largest peak corresponds to the high frequency ultrasonic signal. The side lobes correspond to low frequency modulation. Without the external modulation forces, as shown in the figure, the modulation side lobes disappear. The amplitude of the modulation is related to the interface properties and can be used for quantitative characterization of spring bond density and interface degradation. The amplitude ratio between the carrier at 4.48 MHz and the modulation lobes ±Ω = 18.16 kHz is about 30 dB (the signal to noise ratio is 45 dB).
To illustrate the effect of spring density variations of a layer embedded between two substrates under stress, we have performed stress modulated ultrasonic spectroscopy measurements on as-manufactured (reference) samples and environmentally degraded samples. For each sample, adhesive layer properties and interfacial stiffness have been determined at different levels of stress applied to the bond line. When measurements are performed at a central point of the sample where degradation does not occur, there is no displacement of spectra minima with load variation (for both tension and compression periods of low frequency vibration). This indicates good bond quality. A different situation occurs for points off the sample center as shown in Fig. 6. The ultrasonic reflected signals are collected at different points of the loading cycle. The spectra of the reflected signals are shown in Figs. 6(a), (b) for compression and tension parts of the loading cycle. There is no frequency minima shift during the compression part of the loading cycle (c); however, minimum shift occurs during the tension part. The reflection minimum shifts towards lower frequency with increasing tension load indicating reduction of the interfacial spring. An example of spring reconstruction is shown in Fig. 6(d) indicating interfacial spring stiffness reduction versus tension load. When environmental degradation is much more severe the situation is different. There is no reflection minimum during the tension part of the loading cycle indicating local disbond near the edge (the ultrasonic wave doesn’t show the presence of the adhesive layer). However under compression the disbond closes, establishing good acoustic contact between adhesive and

**FIGURE 6.** Spectra of the reflected ultrasonic signals near the edge of the environmentally degraded adhesively bonded sample under cyclic load. (a, b) Spectra of the reflected signals at different loads: (a) Tension (positive), (b) compression (negative). Shift of the minimum frequency occurs under tension indicating decrease of the interface stiffness under tension and creation of micro disbondings. There is no frequency shift under compression. c) Change of reflection minimum frequency as function of load. d) decrease of interfacial spring during the tension part of the applied loading cycle.
substrate. The ultrasonic signal is reflected from the bond-line with a characteristic minimum, which is shifted toward higher frequency with increase of compression load. The results provided indicate how the stress-modulated method enriches ultrasonic results adding an additional dimension to data understanding and interpretation.

CONCLUSION

A nonlinear stress-modulated ultrasonic method for quantitative characterization of molecular bond density and interface degradation has been developed. It is sensitive to the mechanical bond conditions on the interface under applied load. A model has been developed to describe the non-linear behavior of weak interfaces in adhesive joints and its relation to microstructural strength. Using oblique incidental angle beam ultrasonic spectroscopy we demonstrated that good bonds don’t exhibit reflection minimum shift under vibration load. Interfacial spring stiffness versus load was reconstructed quantifying properties of the bonded interface. Twofold increase of parametric/nonlinear mixing was observed for brittle low strength bonds. It is shown that no difference between this bond and the reference bond can be recorded either by linear angle beam ultrasonic spectroscopy or by a conventional ultrasonic method.

ACKNOWLEDGEMENTS

This work is supported by NASA, Contract # NAS1-00023.

REFERENCES