LASER ULTRASONICS FOR COATING THICKNESS EVALUATION AT 1200°C

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INTRODUCTION

Laser ultrasonics has come of age in such diverse industrial applications as in-process evaluation during steel processing[1] and composite air frame inspection[2,3]. This approach generally offers certain unique advantages for process evaluation and diagnostics. It is a noncontact, largely contour independent, technique ideally suited for hostile environments.

The present application adds to this list high temperature in-process coating evaluation. The hostile Chemical Vapor Deposition (CVD) reactor environment in which silicon nitride coatings are reacted onto carbon/carbon surfaces at 1400°C temperatures as oxidation barriers, in fact, demands the use of remote noncontact methods to extract information such as coating thickness and modulus while deposition is in progress. Such diagnostic techniques can provide direct output real-time parametric data for feedback control of coating deposition. The goal of this effort is to develop laser ultrasound techniques for in-process measurement and control of CVD coating systems.

LASER GENERATION OF ULTRASOUND IN COATING SYSTEMS

Laser generation and detection of waves in free plates has been extensively studied and discussed in the literature. Hutchin's overview[4] is excellent. We shall describe, in this article, the techniques, both experimental and analytic, that were integrated to achieve a successful quantitative evaluation of coating thickness at temperatures up to 1200°C under simulated reactor conditions. This approach will later be applied to an actual reactor under dynamic conditions.

The approach we employed proceeds in four stages. First, laser-generated Lamb waves in free aluminum plates with thicknesses ranging from 75 microns to 750 microns were studied and compared with the literature to perfect the general experimental methods and free plate computer analysis in order to extract the thickness rapidly from the aquired waveform. Next, freestanding crystalline silicon nitride coatings were studied using these methods. The coatings were prepared by removing the carbon on selected samples of silicon nitride that had been deposited upon graphite substrates. The coating is essentially isotropic in-plane since crystal growth is normal. These studies provided new values of sound velocities in this material as well as confirmations of coating moduli necessary for the analysis of the composite system. The third stage was a room temperature study of Lamb wave generation in prepared coatings, ranging in thickness from 50 microns to 200 microns, deposited on a graphite substrate. And, finally, the fourth stage extended the work to high temperatures. Difficulties overcome at each stage prepared the way for the next.

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A major concern in silicon nitride/carbon systems is the avoidance of laser ablation damage either to the coating or to the carbon substrate. This is particularly important at ambient temperatures approaching 1400°C. Laser energy deposition must be low enough not to exceed a phase transition of either material yet acoustic generation must be high enough to obtain adequate signal/noise ratio for an accurate analysis. This was achieved by adopting Cielo's "annulus" technique[5] together with signal averaging. The problem was exacerbated by the crystallinity of the coating which caused severe ultrasonic attenuation via scattering. The annulus approach wherein the YAG laser generation beam is focused into a ring served two functions. It distributed the energy to help avoid ablation while simultaneously focusing one-half the Lamb wave acoustic energy toward the ring center thus amplifying the signal measured by a reciever laser interferometer focused there . It is also important to note that the YAG energy is primarily deposited at the interface since the coating is essentially transparent.

LASER GENERATION AND DETECTION SYSTEM

The system block diagram is shown in Fig.1. A Laser Photonics "Mini YAG" laser provided an 8 ns long, 3mm wide, pulse at a 1 Hz repetition rate. The pulse was expanded to 9mm and sent through an 0.72° positive axicon lens focused to produce an 8mm diameter ring on the target (a negative lens would have been preferable to avoid downstream lens damage). The beam proceeds through a dichroic mirror designed to pass the 1064 nm radiation and 45° reflect the laser interferometer 532 nm beam. The laser interferometer beam was adjusted to be colinear with the YAG source beam upon entering the test furnace chamber. A 500mm target standoff was typical. Careful concentric adjustment of the two beams was crucial to maintain sharply defined wavefronts for analysis. A photodiode triggers waveform aquisition defining t =0. An adjustable delay strip compensates for the electronic delay in the interferometer detection circuit. The waveform is aquired on a LeCroy 9400A digitizing scope. A MacII fx microcomputer operates the digitizer, collects the waveform and analyzes it , all within National Instrument's "Labview" program.

A laser Doppler heterodyne interferometer is used to detect the ultrasonic wave produced by the YAG laser beam.The basic interferometer unit has been described before[6]. The original design was modified for laser ultrasonic evaluation. Fig.2 shows the interferometer block diagram. The source is an 80 mW, single frequency, diode-pumped frequency-doubled YAG laser, ADLAS model DPY315C. The reference and probe beams are both frequency shifted by AOMs. Polarizing prisms are used to direct the light thereby increasing efficiency. Both output beams at the final beamsplitter are detected with front to back 1GHz photodiodes thus obtaining full-wave detection. The diodes detect the 60 MHz difference frequency on the beam which acts as the carrier of the FM Doppler ultrasonic signal. The interferometer beam "turn-on" can be synchronized with the YAG laser pulse by AOM modulation. Since the interferometer is sensitive to the Doppler shift in the light caused by the surface velocity of the travelling ultrasonic wave, it is insensitive to any light intensity changes due, for example, to surface roughness scatter or surface coloration - an advantage with materials like silicon nitrides and carbides. Only 0.05% of the outgoing light intensity need return in order to achieve a useful signal/noise. The interferometer output can be either FM-detected or phase-detected. Phase detection was used for most of the present work.

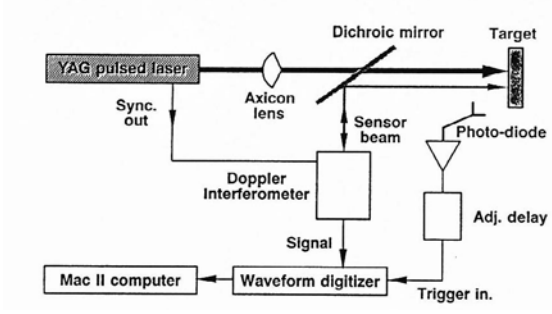

Fig. 1. Block diagram of laser ultrasound system.

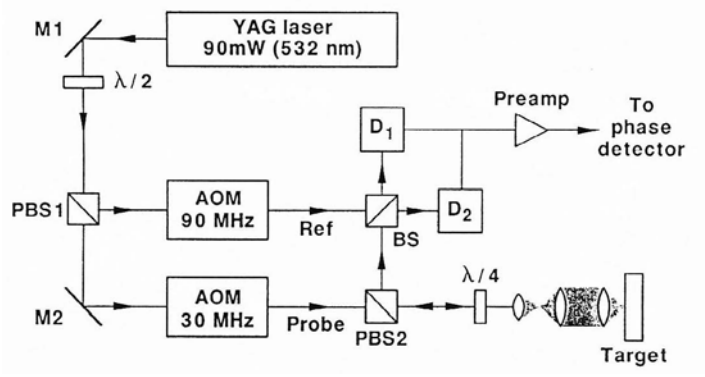

Fig. 2. Heterodyne Doppler interferometer.

Figure 3 shows the Laser Ultrasound system concept diagram with the CVD reactor. Shown also is the use of the axicon lens which generates a ring-imaged YAG laser beam on the component. Thus, with the present concept, a YAG laser pulse will strike the coating surface on a ring generating a Lamb wave which then propagates from the ring boundary inward to the ring center. The laser interferometer, focused at the center, senses the intensely peaked ultrasonic signal resulting from the converging wave thus improving signal/noise ratio while at the same time reducing the risk of ablation. This system will permit remote real time aquisition of ultrasonic data for the determination of coating thickness and coating modulus. A series of laser pulses will sample the coating at regular intervals, for example, once every 15 minutes. In order to minimize the risk of ablation, a low pulse repetition rate is be used together with signal averaging and, of course, the axicon lens. The data is computer analyzed between intervals and outputs a thickness and modulus which can then be used as input for an in-process control system.

FREESTANDING COATING LAMB WAVE ANALYSIS

We have previously stated as a goal the need for real-time analysis since this is an in-process measurement. A brief inspection of the literature will reveal that there has been much analytic effort devoted to Lamb waves in a free plate which is a straightforward analysis, but limited analytic work on leaky Lamb waves in a plate attached to an elastic foundation. Before we develop our approach to leaky Lamb waves, we shall first develop a rapid routine to analyze free plate waveforms and then determine if a similar approach can be found for attached plates. For our free plate analysis we use primarily the analysis of Hutchins [7], and

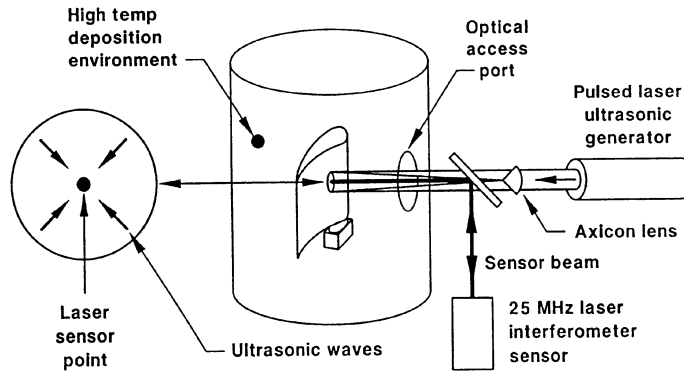

Fig. 3. Laser ultrasound in-process evaluation concept with a CVD reactor.

the approach of Dewhurst and Palmer [8]. Implemented in its full form, an analytic approach based on this work would not meet the real time condition. Specifically, it is desirable to get a coating thickness from an aquired waveform preferably in a matter of seconds but certainly no longer than one or two minutes since thickness data must be aquired every fifteen minutes and it may take minutes for signal averaging multiple waveforms. To achieve this we shall simplify the full analysis by making some plausible approximations in the full theory that enable us to write a simple closed form dispersion relation as a third order polynomial in frequency. The coefficient of each order is then exactly related to the thickness if we specify the membrane velocity which can be experimentally determined. A polynomial fit of the waveform to the dispersion relation allows the extraction of the coefficients and an analytic evaluation of the thickness. In the case of a free plate, the accuracy of fit to all three orders is a few percent and covers a wide range of thickness from 25 microns to 1000 microns (data was taken only over this range) and most certainly beyond.

To begin with, we adopt the approach of ref.[8] to the spectral analysis of the waveform. Very simply, the waveform is scanned for individual components of a well-defined sinusoidal period . The time of flight of that component from the laser strike point is measured thereby obtaining the group velocity and hence a dispersion relation which Dewhurst limits to first order in frequency with fair results. This is a method that can be implemented rapidly on a computer. We then use Hutchins work , stopping short of his phase analysis, to which we apply our approximatons. With these analytic refinements , this general approach results in a thickness evaluation that is more accurate than before allowing one to use, for example, frequencies from 100 kHz or less to 10 MHz or more for the dispersion analysis of thin sheets. A good quadratic or cubic fit is typical.

The dispersion relation to higher order for Lamb waves in a free plate is given by:

$$\omega = A_1 k^2 + A_2 k^4 \quad , \qquad k = \frac{2\pi}{\lambda} \tag{1}$$

The coefficients of the second and fourth order wavevectors are:

$$A_1 = \frac{1}{\sqrt{3}} C_p h \ , \qquad A_2 = \frac{1}{30\sqrt{3}} \left(\frac{20}{r^2} - 27 \right) C_p h \tag{2}$$

C_p is the membrane velocity (also called sheet wave or P-wave) in the plate. It is the symmetric mode and can be measured experimentally as input to this formulation. It can also be found knowing the modulus and density of the plate:

$$C_p = \sqrt{\frac{E}{\rho (1-\nu^2)}} \tag{3}$$

The plate thickness is $2h = \tau$. We shall approximate r, the ratio of the longitudinal to the transverse sound velocity in the infinite medium to be equal to 2.0 . This will be quite accurate for any medium with a Poisson's ratio of approximately 0.33 , that is :

$$r = \frac{C_l}{C_s} \approx 2.0 \tag{4}$$

Using relationships (1) through (4) we find the group velocity, $C_g = \frac{d\omega}{dk}$.

This produces the desired dispersion relation to third order in frequency, f, (using an iterative approximation):

$$C_g^2 = (4a\,\tau)\,\omega + \left(\frac{16b}{a} \tau^2 \right) \omega^2 + \left(\frac{16b^2}{a^3} \tau^3 \right) \omega^3 \tag{5}$$

where, $\omega = 2\pi f$

$a = + 0.2887\ C_p$

$b = - 0.0529\ C_p$

This relationship can now be incorporated in a computer analysis of the aquired free plate waveform in a manner paralleling the Dewhurst approach. For example, the coefficient of the term linear in f is the slope of the squared velocity versus frequency curve used in Ref.[8] to extract the thickness, τ, and reduces to his value.

$$C_g^2 = \frac{4\pi f\tau}{\sqrt{3}} C_p \qquad (6)$$

The computer algorithm can now be described.

COMPUTER ANALYSIS TECHNIQUE

The computer analysis software can be separated into two parts: the first part dealing with the analysis of the plate waves to determine the dispersion of the group velocity, and the second part dealing with the analytic calculation of the plate thickness using the dispersion data.

The group velocity is determined by dividing the distance the plate wave travels by the elapsed time. The elapsed time is defined as the time required for the passing of the peak amplitude in the received plate wave signal. An example of a plate wave signal generated in a 445 micron thick aluminum sheet can be seen in Fig.4a. The membrane velocity in aluminum was taken to be 5.5×10^5 cm/s and travel distance was 2.4 cm between the laser strike and interferometer detection point. Dispersion causes the lower frequency components of the signal to arrive later in time. To determine the group velocity associated with different frequencies, the received signal must first be filtered. The signals that were analyzed for this paper were bandpass filtered using a convolution process. Three-cycle sine and cosine functions modified by a Hanning window were used to define the frequency content of the filtered waveforms. The frequency analysis spans 100 kHz to 1.5 MHz. An example of the modified cosine function for a 500 kHz frequency can be seen in Fig.4b. A convolution of this function with the plate wave signal will yield the portion of the plate wave signal within a 30 percent bandwidth of 500 KHz. The output of the convolution process can be seen in Figs.4c,d for the modulated cosine and sine functions respectively. The two outputs are then squared and summed to form a third time function which is the magnitude of the bandpass filtered plate wave signal. The output for the process can be seen in Fig.4e for the example case. The arrival time of the peak magnitude in this time function is then used to determine the group velocity for the center frequency of the processed signal. By repeating this process over a frequency range, the group velocity dispersion can be measured.

The thickness of a plate can be determined from the relationship between the square of the group velocity and frequency as described previously. To determine the coefficients of the dispersion function, a least-squares third-order polynominal fit was performed on the square of the group velocity data that was measured using the above method. The coefficients of the fit are then equated to the theoretical coefficients (Eq.5) to obtain the plate thickness. An example of the polynomial fit and the dispersion data can be seen in Fig.4f. The three coefficients are shown in Table 1 for several frequency analysis ranges . The 1st order coefficient gives the thickness to an accuracy of 14 % when the maximum 100 kHz to 1.5 MHz range is used and 3 % for 100-600 kHz. The remaining coefficients are not as accurate but confirm the validity of the approximation to higer order and are kept to improve the first order fit coefficient. We use only the first order coefficient generally resulting in free plate thickness errors of less than 5 % for thin plates. As a rule, for a fair analysis, the maximum useful frequency range was determined by deviation from flatness (appearance of a singularity or new mode) at the high end and by deviation from linearity (window width limits) toward the low end.

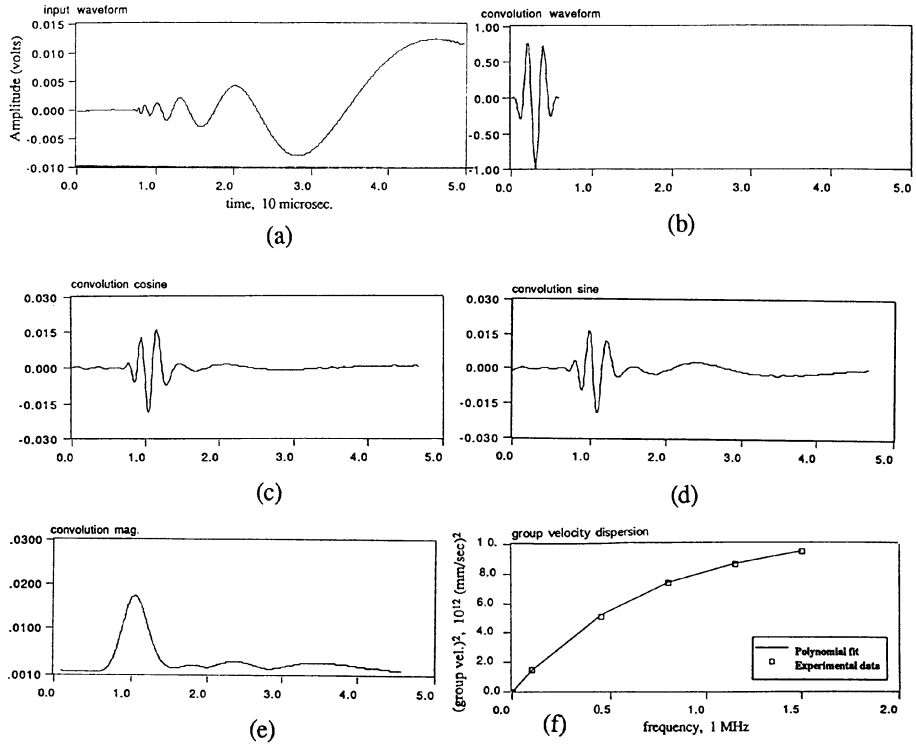

Fig. 4. Computer analysis applied to an aluminum sheet, showing aquired waveform (a), windowed cosine (b), cosine convolution (c), sine convolution (d), convolution magnitude (e), and group velocity dispersion curve (f).

Table 1. FREE PLATE RESULTS - 440 micron thick aluminum sheet

Frequency range (MHz)	0.1--1.5	0.1--1.0	0.1--0.6
1st coef. thickness	381	404	429
2nd coef. thickness	282	315	367
3rd coef. thickness	297	345	437

ATTACHED COATING "SEMI-EMPIRICAL" ANALYSIS

The most common approaches to the problem of leaky Lamb waves on attached plates rely on brute force numerical computer evaluation of the dispersion curves. Perhaps Achenbach[9] comes closest to an analytic evaluation but it has not yet been determined if his approach applies to the present case of a hard coating on a soft substrate in a simple way, permitting a rapid analysis. Due to the pressing demands of the present measurement problem and the difficulty faced in finding a simple closed analytic formulation of the leaky Lamb dispersion relation a semi- empirical approach was attempted.

We have already demonstrated the success of our computer analysis using a high order expansion of the free plate theory. The only input parameter to this approach is the membrane velocity in the freestanding plate. For the present case of a membrane attached to a substrate, a membrane velocity is not as clearcut. There are two limiting mode velocities - that in the coating and that of the substrate. The silicon nitride coating is fast, having a modulus of order 44 million psi while the graphite substrate is extremely slow with a

modulus of only 1.7 million psi. The substrate drags the Lamb wave which penetrates it to a large extent and must slow it down from the measured free plate value. How much is simply a guess at this point. The theory would have to be solved to be more exact. The semi-empirical method simply consists of choosing an "effective membrane velocity", for the attached membrane, intermediate between the substrate Rayleigh speed and the free coating membrane speed,C_p. The "effective membrane velocity" is then to be input to our free plate computer analysis and is adjusted as a single constant that minimizes the thickness error over the largest range of measurements. In the present case, we measured the stated speeds as :

Crystalline Silicon Nitride..... 10.26 x 10^5 cm/s (ρ = 3.18 g/cm^3, ν =0.30)

Graphite..... 1.50 x 10^5 cm/s (ρ = 1.90 g/cm^3)

The graphite Rayleigh speed was measured using the annulus method. The membrane speed in the free coating was measured with a line souce and and with an edge source sending a pulse directly through the plane of the plate. These measurements agreed extremely well. Furthermore, we compared a line source with an annular source in the attached coatings and found the waveforms to be essentially identical deviating only at the very low frequency tail, as expected. To avoid excessive error, the annulus radius should be chosen in proportion to the plate thickness and should be several times the longest wavelength expected. For example, for our coatings, the longest resolved wavelength is 1- 2 mm. A radius of at least 3 - 6 mm is desirable. Our choice here was unfortunately also constrained by the 5 mm radius of the reactor optical access port .

ROOM TEMPERATURE COATING THICKNESS RESULTS

Waveforms were collected on 3.8 cm x 3.2 cm , 0.5 cm thick, graphite bars coated with nominally 50 microns, 100 microns and 200 microns of crystalline silicon nitride. A YAG beam annulus radius of 3.21 mm was chosen. An annulus thickness of 0.5 mm for energy distribution was achieved by defocusing. Figures 5-7 show the aquired waveforms after 100 averages and the dispersion curve for each waveform. The waveform of figure 6 was offset to eliminate the echo from the spectral analysis and actually begins at t = 0.5 microseconds. Thickness results obtained from our semi-empirical analysis are compared to the measured thickness in table 2. We found that an intermediate " membrane velocity" value of 6.32 x 10^5 cm/s, when introduced into the free coating program, produced less than 10% error in the thickness measurement of the crystalline attached coating over the full range of data from 50 microns to 200 microns taken at room temperature. This surprisingly good fit suggests that velocity "mixing" or averaging may provide a simple representation to achieve some sort of an approximation for the theory. In reality, one expects that the "effective velocity" should be a function of the frequency as well, but we apparently find that it is a relatively weak function. We then use this value in all our analyses for this coating system at room temperature. The question naturally arises as to whether these assumptions are valid at high temperatures. It will work as long as there is no significant temperature dependence of the dominant modulus, namely the coating. The influence of the substrate is not clear. We undertook a series of waveform measurements from room temperature to 1200°C partly to ascertain the accuracy of these assumptions at high temperatures.

HIGH TEMPERATURE COATING THICKNESS RESULTS

For the high temperature work, a resistive furnace with a 3-inch bore was employed to heat the specimen up to 1200°C. A quartz tube fitted with water cooled and insulated end flanges, an optically flat laser access window on one flange, and an inert gas flow system, was used to contain the coated graphite bar which was mounted on a quartz support at the tube center. The sample was allowed to equilibrate one-half hour at each temperature. An average of 100 waveforms was taken. Reproducibility was checked at several different points on the bar and found to be well within the error of the theory and experiment. 10 mJ of YAG pulse energy at a 1 Hz repetition rate was employed and distributed over a 3.75 mm radius, 0.5 mm thick annulus. The interferometer beam was carefully aligned at its center.

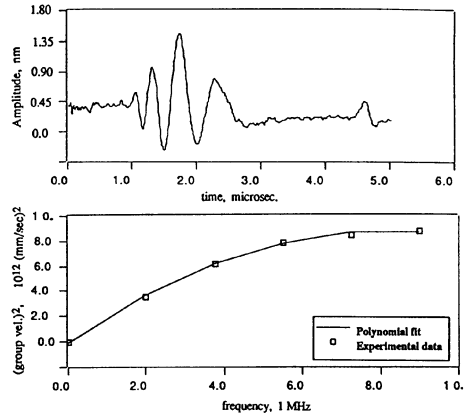

Fig. 5. Aquired Lamb wave(above) and group velocity dispersion curve(below) in 50 μ Si_3N_4 coating on graphite.

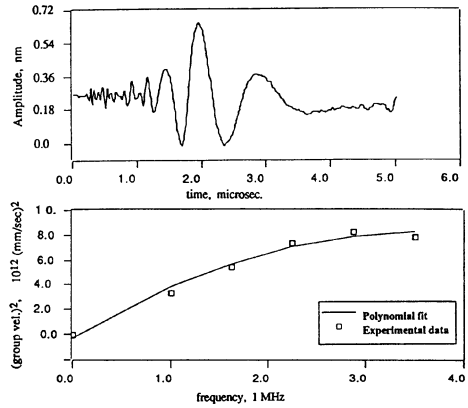

Fig. 6. Aquired Lamb wave(above) and group velocity dispersion curve(below) in 100 μ Si_3N_4 coating on graphite.

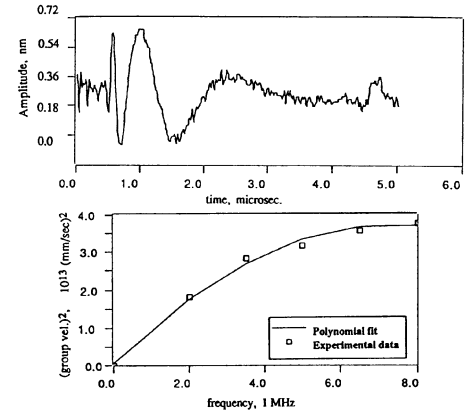

Fig. 7. Aquired Lamb wave(above) and group velocity dispersion curve(below) in 200 μ Si_3N_4 coating on graphite.

Table 2. ATTACHED COATING RESULTS - Crystalline Si_3N_4 on graphite

Actual thickness (microns)	Calculated Thickness
50.5	46.4
98.0	102.1
208.3	214.4

Waveform data are presented in Fig. 8 for a 50 micron crystalline silicon nitride coating on a graphite substrate from room temperature to 1200°C. As is evident,there is no significant change in shape over this range. Indeed the signal/noise is essentially constant indicating no significant change in optical reflectivity or ultrasonic attenuation as well. A P-wave is visible near the front of the waveform apparently moving at the free coating membrame velocity. In addition, there is a changing echo from the back of the bar. This echo is the forward longitudinal pulse arising from stress enhancement of the YAG energy conversion at the graphite/nitride interface. The round trip echo time apparently decreases with increasing temperature to 1200°C indicating a corresponding velocity increase. This is consistent with a known increase in graphite modulus with temperature up to 1200°C. Hot gas convection in the tube at 1200°C generated optical fluctuations (weak scintillations - like starlight) in the interferometer but had no effect on the data because of its Doppler sensitivity. Figure 9 summarizes the thickness analysis of these waveforms over the given temperature range for the single 50 micron coated specimen. The same "effective membrane velocity" of 6.32 x 10^5 cm/s was used in the program as was used at room temperature. The constant 53 micron thickness calculation over the entire range suggests that any velocity change with temperature is well within our stated 5% stated error for thin coatings.

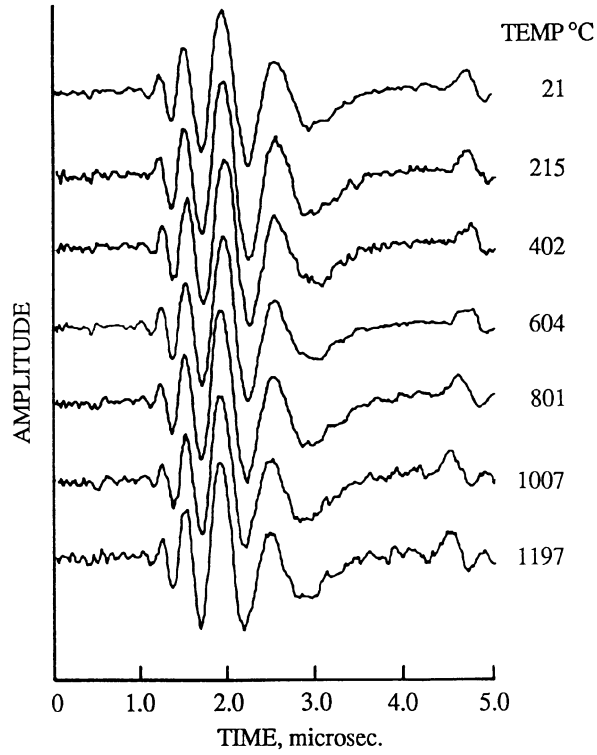

Fig. 8. Lamb wave as a function of temperature in 50 μ Si_3N_4 coating on graphite.

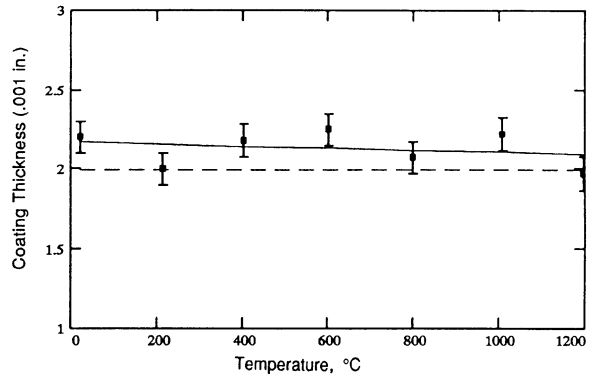

Fig. 9. Thickness as a function of temperature in 50 μ Si_3N_4 coating on graphite.

CONCLUSIONS

We have shown that laser ultrasound is a fruitful approach to the quantitative measurement of physical properties of coatings up to 1200°C. The standard free plate analysis of coating thickness has been extended to include hard plates attached to soft substrates using a "semi-empirical" approach. This problem will be addressed analytically to confirm our observations. It is expected that this approach should also work in the dynamic atmosphere of a CVD reactor to 1400°C without significant modification.

ACKNOWLEDGEMENTS

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