

IN-SITU HIGH-TEMPERATURE MONITORING OF CARBON-CARBON OXIDATION USING TIME-REVERSAL MIRRORS

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ABSTRACT

The development of methods for in-situ monitoring of high temperature composites materials during oxidation is important in a number of technological applications. This work is the development of methods that make use of solid cylindrical multi-mode waveguides to monitor the oxidation reaction in carbon-carbon composites.

A time reversal mirror process was used with a solid circular cylindrical waveguide to remove the effects of dispersion. The first three longitudinal modes were considered. An analytic transfer function was developed to predict the time-reversed signal from an input signal. The analytic transfer function was applied to the time-reversed signal to reproduce the original input signal. Experiments were then performed to demonstrate the fidelity of the analytical model.

Results of the comparison were reasonable and showed the potential of the approach for use with a high temperature ultrasonic buffer rod. Results are shown for preliminary coupling studies that will be used to help in characterization of ultrasonic attenuation and the increase in the accuracy of the time reversal mirror. A signal obtained during an oxidation of carbon composites is also shown. Future work will focus on the better integration of the time reversal technique to obtain quantitative information from the experimental apparatus described. Applicability to other material systems and measurements will also be discussed.

MOTIVATION

The primary objective of the work described is the development of techniques to measure the in-situ characteristics of composites at temperatures above 1000°C. A number of important civilian and military applications for composites exist in these high temperature regimes. However the cost effectiveness depends on highly optimized design and durability models for the material. Of course the anisotropy must be exploited in order to achieve the desired performances as well in a number of these applications. However, while the room temperature design process is reasonably well understood and has shown great progress in recent years, the degradation of the matrix and the selective diffusion of oxygen in many of these materials are less well understood. At least a portion of this lack of understating is due to the absence of in-situ sensing techniques that can allow the process dynamics of the degradation to be monitored. Optimization for complex composite structures where load paths may vary requires knowledge of the off-axis elastic and strength properties as well as the changes in these properties during oxidation processes. Applications for these methods would include evaluation of carbon-carbon and other materials for use in oxidizing environments that exist in missile defense applications and developing increased understanding of the durability of these materials for the civilian space program. The technique

is not specific to carbon systems, since similar mechanisms may also exist in other emerging materials for high temperature applications. The emphasis is placed on methods that can be used to recover the full constitutive tensor [C] as well as the changes that would occur during oxidation. In-situ methods will allow repeatability studies to be performed to assess sample-to-sample and test-to-test variability without creating an unreasonable test matrix for the material.

BACKGROUND

In a number of applications a need exists for techniques to determine a complete set of elastic anisotropic material properties at high temperatures. A particular application is for high accuracy measurements of the elastic properties as a function of temperature and as a function of the rate of temperature change. However, because of limitations in the size and quantity of specimens available and limitations in the availability of testing equipment, significant obstacles remain. New methods need to be developed where appropriate and the results of the methods should be compared to other documented results.

Elastic waves have the advantage of providing a non-destructive method of evaluating the elastic properties, in particular the modulus and density, of an anisotropic material. The number of propagating modes and the shape of the dispersion curve can be used to simultaneously determine a number of the material properties for an anisotropic material. The proposed method differs from other efforts in this area in the use of a reconstruction algorithm for the characterization of dispersive modes. By making use of the dispersion of the modes along with the changes in the group and phase velocity, as well as the attenuation of the modes, fully anisotropic, linear visco-elastic characterization of materials is possible in-situ. Shear and Young's modulus measurements are possible in-situ from the evaluation of a single received signal [1]. This approach has been shown in previous studies to yield extensive information on the nature of a disturbance that has propagated through either a dispersive material or through a dispersive geometry [2, 3]. Additional applications in this area have shown the potential for waveguide sensors to provide real-time information in process sensing and materials research applications [4, 5, 6].

The determination of the elastic properties of a material is attractive because of the potential of this method to characterize the degradation of properties of a composite after time at an elevated temperature or the change in material properties as a function of temperature. In particular damping changes (attenuation of the elastic wave) are quite sensitive to small levels of change in the matrix of the composites. The approach that has been taken is elastic waveguide monitoring. While acousto-optical methods are attractive for some applications, elastic waveguides are useful in some cases because of the insensitivity of the technique to changes in surface reflectivity. However, acousto-optic sensing has shown promise in some applications where this is not an issue. For example, acousto-optics has been successfully applied to the monitoring of the sintering of zinc oxide [7].

Multi-mode waveguides are a low cost and effective method of providing an acceptable signal to noise ratio at high temperatures [6]. Dry coupling of the buffer rods to the sample has been shown to be possible by a slight modification of the standard technique. While the complexity of the received signal is high (the waveguide propagates multiple waveguide modes which are not separated in time) the signal to noise ratio is reasonable. High temperature waveguide buffer rods not only show great promise for overcoming the limitations on cost and complexity, they also have the potential to provide important additional information which would be difficult to acquire using acousto-optical methods. By exploiting the dispersive nature of higher order waveguide modes, and using multiple waveguide modes, it is also possible to separate out the geometrical changes from the changes in material properties. Using the waveguide modes could also produce an image of the cross sectional density gradient of the sample. Reconstruction of an in-situ cross-sectional image of the material would make it possible to track the effects of the diffusion controlling parameters versus time at temperature. Heat flux controlled degradation of a composite would be an example of such a process. For simple elastic property measurements below the material degradation temperature, elastic properties can be measured at a range of temperatures without risk of damage to the specimen.

By generating more data during each test run, the effect of sample dependent conditions can also be significantly reduced, further reducing the number of measurements required. This real-time data could either be used to determine times at which the samples need to be removed for strength testing or to determine if the bulk properties of the sample have been affected by holding the sample at a high temperatures for shorter time periods.

Related work considers applications for waveguides for the monitoring of die casting [8] as well as the previously mentioned work in reaction bonding of silicon nitride [6]. The other work suggests that the sensor developed in this work may be applicable to the monitoring of related problems in ceramic processing as well as in the monitoring of physical and chemical vapor deposition processes. Cross-sectional density gradient monitoring is of particular importance in applications such as the production of carbon-carbon composites as well as in filling of high aspect ratio vias in chemical vapor deposition processes in the electronics industry.

EXPERIMENTAL APPARATUS

The basic apparatus used in this work consists of a tube furnace with long multi-mode cylindrical waveguides that isolate the piezo-electric transducers for the high temperature environment in the furnace. The entire apparatus is controlled so that a cooling system protects the transducers and the environment can be modified to move the sample from an inert to an oxidizing environment. The furnace employed for this work is a conventional tube type furnace (Lindberg Model 55346, Watertown WI) with heating elements capable of producing sample temperatures of approximately 1100°C. The furnace used is a 3-zone furnace, however for the experiments performed the outer zones were not observed to differ significantly from the central zone regardless of settings. The furnace is evacuated using a medium vacuum pump (Marvac Scientific, Model #R-10, Concord CA) that is attached to the sealed furnace tube. The apparatus is also supplied with an inert gas atmosphere (argon) from bottled source. Nitrogen is used to purge the furnace prior to performing the tests in an oxidizing environment. The system was purged with the vacuum pump and then back filled with nitrogen. This process was repeated several times in each experiment to eliminate oxygen from the furnace environment. Argon is then used during the heating of the sample. Finally, once the sample is at equilibrium at temperature medical air is flowed through the furnace to provide a controlled oxidizing environment. All experiments are performed at a slight pressure about atmospheric. The pressurized furnace maintains the atmosphere even in the presence of leaks. A gas flow meter is used on the output of the pressure tank to monitor leakage from the system. Simultaneous measurement of the furnace temperature profile on the buffer rods is also possible for up to eight channels using a switchable thermocouple system.

The ultrasonic system used is a conventional pulser with standard low temperature transducers. The initial waveguide experiments were carried out using a spike pulser (Panametrics Model 5072PR, Waltham MA). For the higher amplitude required for monitoring samples in the furnace a square wave pulser is used (Ritec SSP-801, Warwick, RI). The receiving transducer uses a separate pre-amp (Panametrics 5660C) to improve the signal isolation due to low signal to noise ratio in the received signal. The transducers used are broadband nominal 1 MHz. central frequency units (Panametrics V103, Waltham MA) that are coupled to the end of the waveguides using High Vacuum Grease (Dow-Corning, Midland MI). Standard room temperature transducers are used since most "high temperature" transducers are significantly past their Curie temperatures at the temperatures of interest in these experiments. High temperature transducers are also significantly less efficient than the room temperature counterparts. In this application large signal amplitudes are required because of the attenuation that occurs in the sample, in the coupling between the buffer rods and the sample, and in the coupling between the buffer rods and the transducer. The cooling system associated with the waveguides is then sized to maintain the transducers at the lower temperature.

Thus the key to the apparatus is the design of the end-caps used in the system. The end cap configuration is shown in an exploded view in Figure 1. The ultrasonic transducer is designed to maintain continuous

contact with the end of the waveguide. The waveguide and the transducers are withdrawn when measurements are not being made on the specimen in the furnace. Small, one-inch diameter air cylinders, that are operated using the inert gas, are used to retract and extend the waveguides. A series of water-cooling coils are wrapped around a double set of ball bearings. These ball bearings are the only contact between the furnace frame and the waveguides. This design allows the waveguides to be retracted as well as being cooled by the water jacket. The entire system is air tight, with the only penetrations of the furnace end caps used for the ultrasonic cable and the thermocouples and other wiring. A spring-loaded fixture that holds the transducer provides constant pressure between the transducer and the waveguide.

Unlike the intermittent contact between the buffer rods and the sample, the buffer rods and transducers can be in continuous contact. In order to restrict movement of the transducers, they are clamped along with the buffer rods in a cylindrical housing. Set screws are used to secure the transducer and buffer rod into place. The quartz buffer rods however prove to be a bit more difficult due to the brittle nature of the quartz. Minimal pressure applied to the setscrews along with the use of a Nylon tip will decrease the chance of damaging the buffer rods.

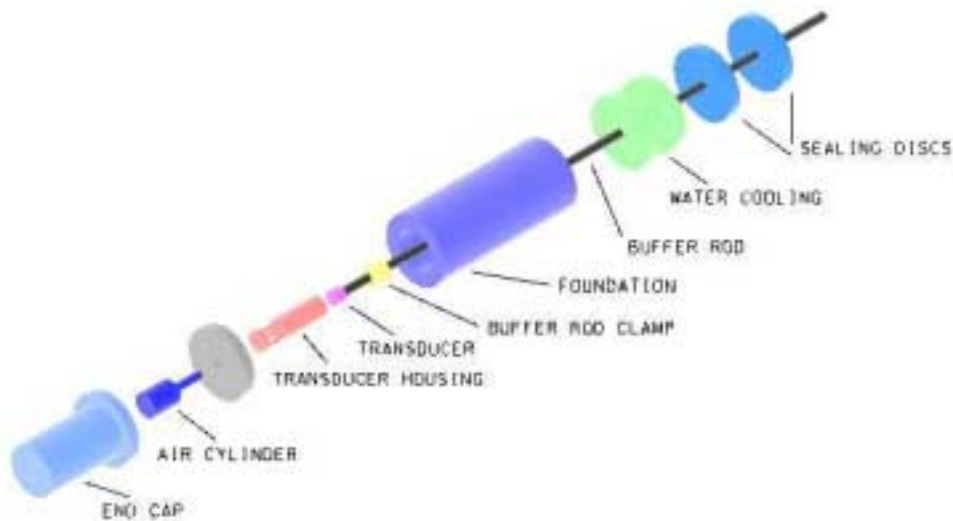


FIGURE 1. An exploded view shows the end cap and frame for housing the transducer, air cylinder, and buffer rod. The cooling system is also shown.

EXPERIMENTAL FURNACE RESULTS

Intermittent contact between the sample and the buffer rods is used for several reasons. First, the buffer rod inhibits the flow of the atmosphere around the sample during the testing. When the buffer rod is withdrawn the influence on the atmosphere is reduced and any cooling associated with the contact of the buffer rod with the face of the sample is eliminated. Withdrawing the buffer rod also reduces contamination of the end of the buffer rod by material from the sample. Even heating of the end of the buffer rod is also ensured. At the higher temperatures used in this testing the fused quartz buffer rods approach their softening point, which significantly changes the properties of the material. The softening may increase the coupling between the buffer rod and the sample. Finally, the buffer rod is withdrawn so that the ultrasonic signal from the free end of the buffer rod may be acquired. This signal is used in the deconvolution of the signal. The effects of changes in the rest of the system make it necessary to obtain a number of signals in order to extract the material response of the sample (Peterson, 1994).

The configuration of the carbon samples and the alumina support tooling is shown in Figure 2. This configuration allows gas flow around the sample and withdrawal of the buffer rods during times at which measurements are not being taken. The buffer rods are shown in both a contact and a withdrawn configuration. An example of a signal that has been propagated through a carbon sample at 1000°C is shown in Figure 4. The signal to noise ratio in the signal is quite good and shows the effects of the complex signal which results from propagation through waveguides that propagate multiple modes.

TIME REVERSAL MIRRORS

As is evident from figure 3, the significant complexity of the signal makes measurements of arrival time quite difficult. While this is true for the through transmission configuration, it is even more true for the reflected signal that is used in the deconvolution [6]. Thus, a method is desirable that would allow a simpler signal to be used in the processing of the data.

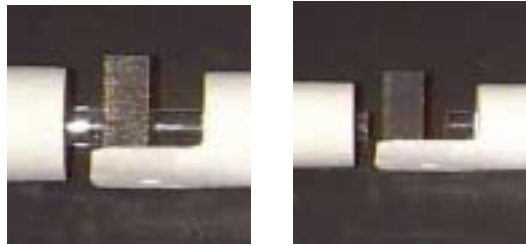


FIGURE 2. Photographs show the intermittent contact between C/C sample and Quartz buffer rods.

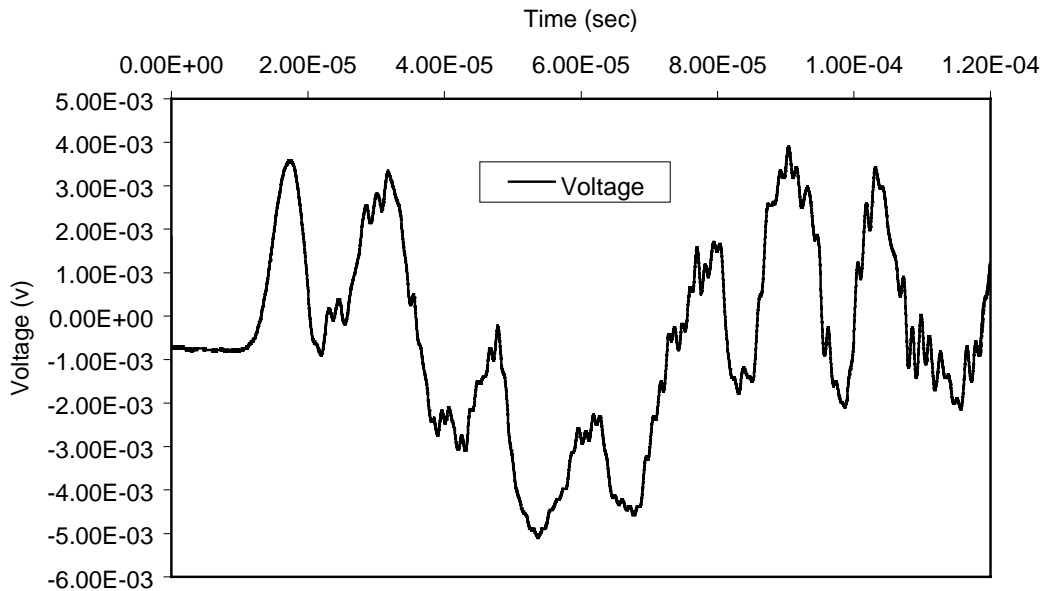


FIGURE 3. Received signal after propagating through the Quartz buffer rods at 1000°C.

In a thick waveguide the acoustic signal propagates along multiple paths that superpose to create propagating modes in the waveguide. The propagation of multiple modes causes a signal that is compact in the time domain to have a large time signature after propagating through the waveguide, Figure 4 shows both the input and the transmitted signal for the a solid waveguide used similar to that use in the furnace application. As a result, information obtained from changes is harder to extract. A number of approaches have been considered to solve this problem, however the complexity of accommodating the multi-mode signal is high [1]. A time reversal mirror (TRM) has been shown to eliminate this problem in a through transmission configuration in a cylindrical waveguide by removing the complexity of the signal [9].

The use of a TRM in a pulse-echo configuration with a solid cylindrical waveguide is shown as an example. This corresponds to one of the six signals used in the deconvolution for the data from the furnace. Signals were recorded both with and without a sample on the end of the waveguide. The TRM was able to produce a signal with a compact time signature in both cases. Information about the aluminum sample was obtained in a comparison between the two signals.

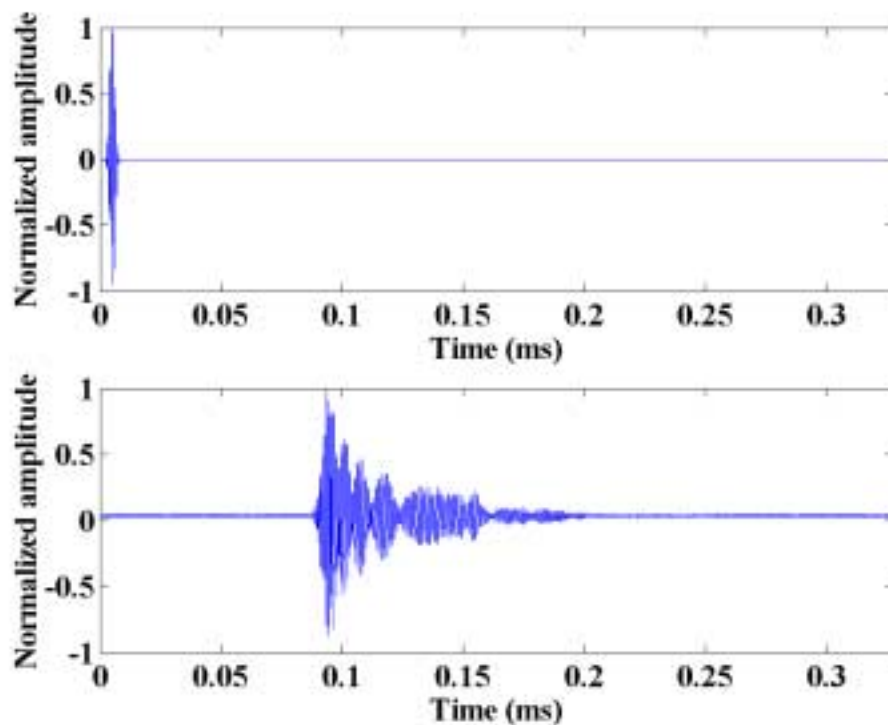


Figure 4. Illustration of dispersion in the cylindrical waveguide showing the loss of compact support.

TIME REVERSAL MIRRORS

A time-reversal mirror experiment consists of three steps. In the case of a cylindrical rod, first, an acoustic signal is excited by a source at one end of the rod. The acoustic signal propagates through the rod, and the altered signal is recorded at the opposite end. Second, the recorded signal is reversed in time. Finally, the receiver is excited with the reversed signal. The reversed signal propagates through the rod, and a new signal is recorded at the source. If time invariance is satisfied this new signal is the same as the original acoustic signal except reversed in time. The ability of the TRM to determine the input signal needed to produce a desired output signal can produce a compact time signal from a dispersive system.

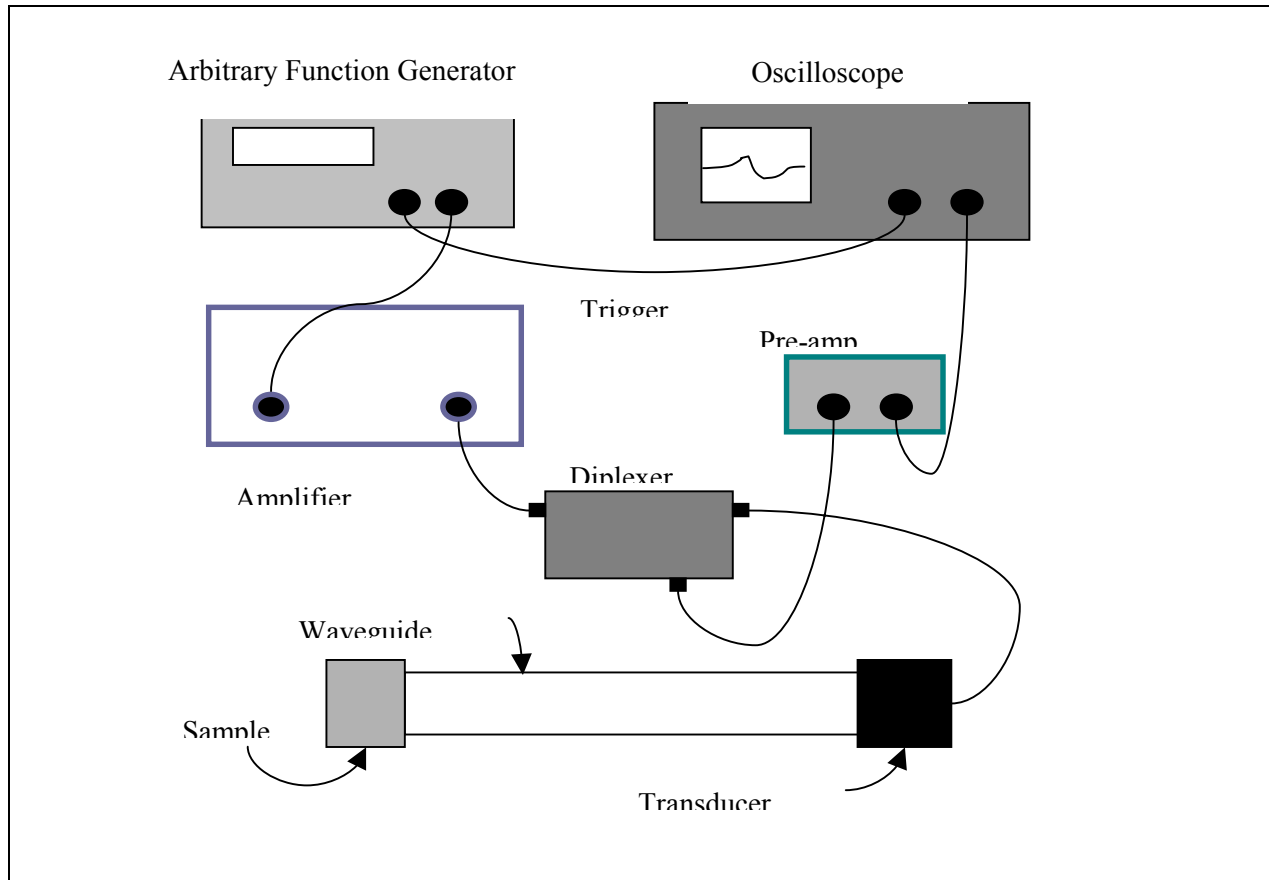


Figure 5. Diagram of the experimental setup for time reversal mirror.

The pulse echo configuration used for the experiments is shown in Figure 5. The waveguide consisted of a fused quartz cylindrical rod, 228 mm in length. Fused quartz has a Young's modulus, E , of 73 GPa, a density, ρ , of 2200 kg/m³, and a Poisson's ratio, ν , of 0.14. The data shown is for a waveguide that is larger than that which is used in the furnace, however the technique is applicable to the smaller diameter waveguide as well.

The pulse-echo configuration uses a single transducer that acts as the source and the receiver. The transducer used in the experiment was a 38 mm diameter, 1 MHz broadband longitudinal contact transducer [Panametrics, model V194, Waltham, MA]. A coupling fluid was used between the transducer and the waveguide and between the waveguide and the sample [Sonotech, Inc. UT-30, State College, PA]. A pulser [Panametrics, 5072PR, Waltham, MA] was used to generate a pulse to the transducer. The dispersed signal was recorded and reversed in time. An arbitrary waveform generator [Agilent 33250A, Palo Alto, CA] produced the time-reversed signal to drive the transducer. A radio frequency power amplifier [ENI A-300, Rochester, NY] with a gain of 55 dB was used to amplify the signal to the transducer. The received signal was recorded by a digital storage oscilloscope [Tektronix TDS 520A, Wilsonville, OR] after amplification of the signal by an ultrasonic pre-amplifier [Panametrics, model 5660, Waltham, MA] with a gain of 40 dB. In order to use the transducer in pulse-echo mode with the arbitrary waveform generator a transformer diplexer [Ritec Inc., model RDX2, Warwick, RI] was placed between the transducer, the ultrasonic pre-amp, and the power amplifier.

The TRM proved to be effective in the pulse echo configuration. The time-reversed signal was used to excite the transducer, and the echoed signal received by the transducer was a pulse, top graph in Figure 3. The most important characteristic of this experimental signal is the compact time signature. By using the time-reversed signal as the excitation signal, the dispersive properties of the waveguide can be negated.

This capability allows the use of a dispersive solid circular waveguide as a low cost sensor. The compact time domain signal greatly simplifies signal analysis that was previously used [6].

To explore the ability of this technique as a sensor a 25.4 mm aluminum cube was placed at the free end of the waveguide. The same time-reversed signal was used to excite the transducer. The received signal includes both front and back wall reflections from the aluminum cube, bottom graph Figure 3. The first peak corresponding to the reflection at the end of the waveguide was attenuated compared to the peak from the reflection from the free end of the waveguide. The attenuation results from transmission into the finite impedance material. Also a second peak was generated from the reflection of the back wall of the sample. The time delay between the two peaks correlates to the bulk wave speed in aluminum and the thickness of the cube.

From these experiments the technique is promising as a means to detect changes in impedance or wave speed in an actual application. For a practical application with a single waveguide, the signal that will cancel the dispersive effects of the waveguide is easily determined from the TRM experiment. For more complex configurations where significant changes with time are expected either modeling or more extensive experiments are required [10]. Future work remains to be done to show that measurements can be made in-situ and to develop appropriate models.

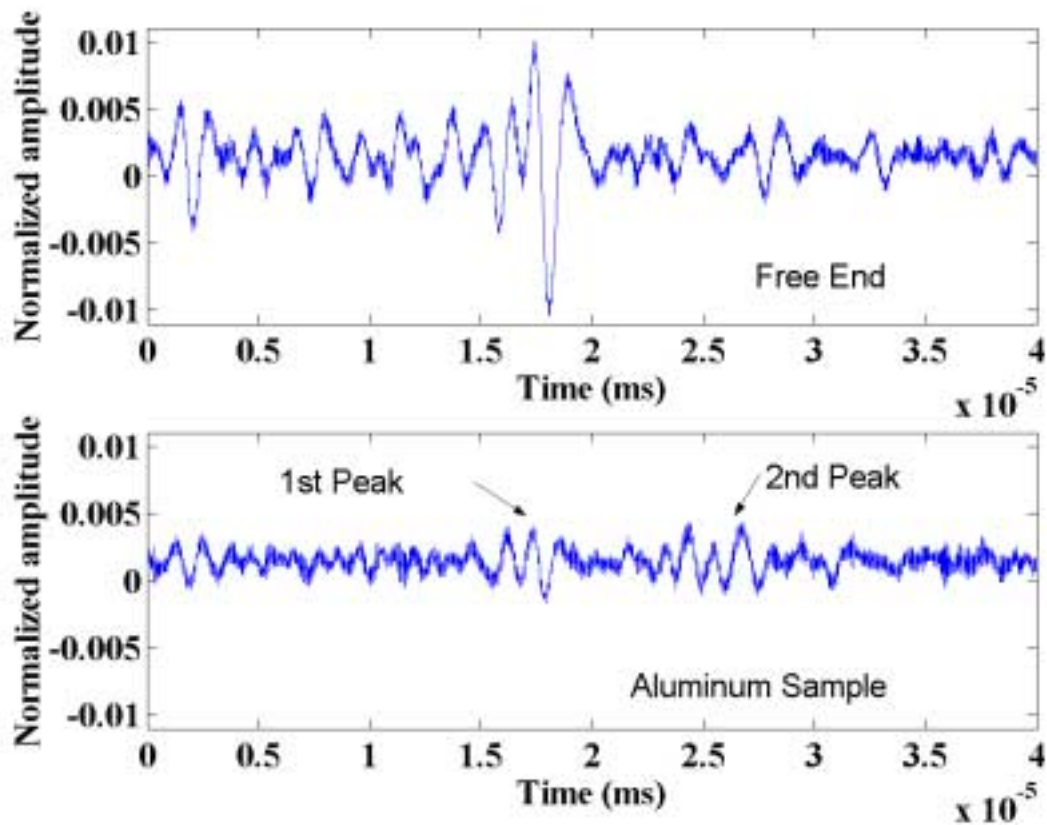


Figure 6. Comparison of received signals.

CONCLUSIONS

Results have been shown for the use of the thick solid cylindrical waveguides for the in-situ monitoring of the oxidation of carbon composites. The results demonstrate that this type of technique is useful for these measurements and that measurements of the samples are possible at temperatures up to about 1000°C. However, the results also have shown that the signals that are acquired from the approach are quite complex. In the past extensive signal processing has been used to analyze the data. An alternative approach is shown that uses time reversal mirrors to produce simpler signals from the ultrasonic waveguides. This approach has the potential to produce a signal that is comparable to a simple experimental apparatus in a large and complex system such as the one shown. Future work will make use of multiple time reversal mirrors to produce a set of six signals that are both simple to process and are usable for an in-situ tests system such as the one shown.

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