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Update on Ultrasonic Thermometry Development at Idaho National Laboratory

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ABSTRACT

The Idaho National Laboratory (INL) has initiated an effort to evaluate the viability of using ultrasonic thermometry technology as an improved sensor for detecting temperature during irradiation testing of advanced fuels proposed within the Fuel Cycle Research and Development (FCR&D) program sponsored by the U.S. Department of Energy (US DOE). Ultrasonic thermometers (UTs) work on the principle that the speed at which sound travels through a material (acoustic velocity) is dependent on the temperature of the material. UTs have several advantages over other types of temperature sensors . UTs can be made very small, as the sensor consists only of a small diameter rod which may or may not require a sheath. Measurements may be made up to very high temperature (near the melting point of the sensor material) and, as no electrical insulation is required, shunting effects observed in traditional high temperature thermocouple applications are avoided. Most attractive, however, is the ability to introduce multiple acoustic discontinuities into the sensor, as this enables temperature profiling with a single sensor. The current paper presents initial results from FCR&D UT development efforts. These developments include improved methods for fabricating magnetostrictive transducers and joining them to waveguides, characterization of candidate sensor materials appropriate for use in FCR&D fuels irradiations (both ceramic fuels in inert gas and sodium bonded metallic fuels), enhanced signal processing techniques, and tests to determine potential accuracy and resolution.

Key Words: In-Pile, Ultrasonic Thermometry

1 INTRODUCTION

The Fuel Cycle Research and Development (FCR&D) program emphasizes an approach relying on first principle models to develop optimized fuel designs that offer significant improvements over current fuels. To facilitate this approach, high fidelity, real-time, data are essential for characterizing the performance of new fuels during irradiation testing. The fuels of interest to the FCR&D program pose several challenges in terms of temperature monitoring. Ceramic fuels (e.g. uranium dioxide) for fast reactor applications may be tested with pellet centerline temperatures of up to 2600 ^{o}C. Metallic fuels (i.e. uranium alloys) are tested with lower pellet centerline temperatures (peak values of 1100 ^{o}C), but these fuels are bonded to liquid sodium or liquid metals (both highly corrosive) to enhance heat transfer.

Ultrasonic thermometry has the potential to improve upon temperature sensors currently used for in-pile fuel temperature measurements. Current methods for in-pile temperature detection primarily rely on either thermocouples or post-irradiation examination methods (such as melt wires). Commercially-available thermocouples (e.g., Type K, Type N, Type C, etc.) are widely used and cover a wide temperature range. However their use is limited. Type K and Type N thermocouples decalibrate at temperatures in excess of 1100 ^{o}C. Material transmutation causes decalibration in tungsten/rhenium (e.g., Type C) or platinum/rhodium (e.g., Type R or S) thermocouples in neutron radiation environments. Even though the High Temperature Irradiation Resistant Thermocouple (HTIR-TC) developed by the Idaho National Laboratory (INL) has overcome most of the difficulties associated with thermocouples [1], the resistivity of electrical insulators can degrade if subjected to high temperatures (in excess of 1800 ^{o}C), causing shunting errors. Although larger diameter, multipoint thermocouples are available, most thermocouples only measure temperature at a single location. Melt wires and other post irradiation methods only allow estimation of maximum test temperatures at the point of installation. The labor and time to remove, examine, and return (if necessary) irradiated samples for each measurement also makes this out-of-pile approach very expensive. Prior ultrasonic thermometry applications have demonstrated the viability of this technology, but in-pile applications were primarily limited to high-temperature fuel damage tests, which ceased several decades ago [2].

Ultrasonic Thermometers (UTs) work on the principle that the speed at which sound travels through a material (acoustic velocity) is dependent on the temperature of the material. The most studied (and, therefore, most well-developed) form of ultrasonic thermometry is the pulse echo method. The average acoustic velocity (and, therefore, the average temperature of the rod) is calculated by sending an ultrasonic pulse through a thin rod of known length and measuring the time between the initial pulse and the reflection of the pulse from the opposite end of the rod. By introducing acoustic discontinuities such as notches or sudden diameter changes into the rod, the probe may be segmented into a multipoint temperature sensor (the average temperature of each segment derived from timing of the successive reflections). In order to avoid wave dispersion effects, the rod should have a diameter of less than one tenth of the signal wavelength [3]. If this condition is met, the temperature-dependant acoustic velocity of the sensor material, $c(T)$, is related to density, $\rho(T)$, and elastic modulus, $E(T)$, (both properties are also temperature dependant) of the sensor material through the following equation:

$$c(T) = \sqrt{\frac{E(T)}{\rho(T)}} \tag{1}$$

A typical multi-sensor UT system, with key components identified, is shown in Figure 1. As indicated in this figure, a narrow ultrasonic pulse is generated in a magnetostrictive rod by a short duration magnetic field pulse produced by an excitation coil. The ultrasonic pulse propagates to the sensor wire, where a fraction of the pulse energy is reflected at each discontinuity (notches or diameter change). Each reflected pulse is received by the excitation coil, transformed into an electrical signal, amplified and evaluated in a start/stop counter system. The time interval between two adjacent echoes is evaluated and compared to a calibration curve to give the average temperature in the corresponding sensor segment. When a number of notches are available on the wire sensor, the various delay time measurements give access to a temperature profile along the probe.

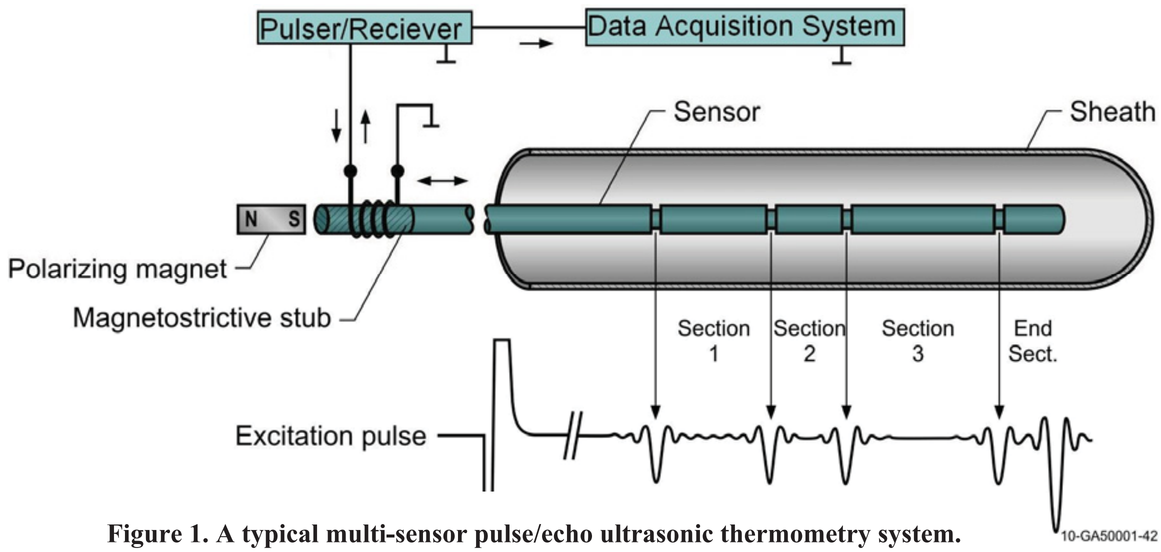

Figure 1. A typical multi-sensor pulse/echo ultrasonic thermometry system.

In order to develop UTs for fuel and material irradiation tests, appropriate candidate sensor materials must be identified and evaluated. Sensor materials must have high melting temperatures, low thermal neutron absorption cross sections, and resist interactions with the fuels or fill materials that they will be in contact with during an irradiation test. The materials must have an appropriate temperature response. This paper presents results from initial testing that was completed to evaluate the temperature response of several candidate sensor materials. Section 2 describes the overall process that will be followed to demonstrate the viability of a UT for irradiation testing. This section also presents the setup used for initial acoustic velocity testing and the process used to select and fabricate sensor samples for these tests. Section 3 presents results from these initial tests. Section 4 summarizes results and summarizes the remaining steps required before UT sensors will be available to support irradiation testing.

2 ACOUSTIC VELOCITY CHARACTERIZATION

Development of a viable UT system will require improvements to several aspects of the technology. First, appropriate materials will need to be identified which can survive harsh environments without significant loss of performance over the duration of an irradiation test. Next, an improved method of processing signals from the thermometer will be developed. These signals become very complex when multiple sensor segments are required, as reflection signals can overlap each other. This problem can be compounded by "sticking;" contact bonding between the sensor and its surroundings. Sticking causes extraneous reflections at very high temperatures which can completely obscure the sensor signals. A method will be developed to eliminate sticking. Finally, a prototype system will be developed and tested to assess the potential accuracy and resolution that a UT system may achieve.

As an initial step in the UT system development effort, acoustic velocity characterization tests were completed in order to reduce the number of candidate materials by comparing temperature response, ease of fabrication, transmitted signal quality, etc. The tests were also used to evaluate and make appropriate adjustments to a signal processing technique for UT applications. The steps required to perform these tests (candidate material selection, test sample fabrication, test setup, and signal processing adaptation) are detailed in the following sections.

2.1 Sample Material Selection

Candidate sensor materials were selected based on melting temperature, thermal neutron capture cross section, and compatibility with likely in-pile test conditions. For higher temperature applications, such as inert gas-filled tests of oxide fuels, refractory metals are an obvious choice due to their high melting temperatures. Some refractory metals have been used successfully in similar tests for evaluating thermocouple components (both for short term use in tungsten-rhenium thermocouples and for longer term use in molybdenum-niobium based HTIR-TCs developed at INL). Tungsten and rhenium were not considered (despite high melting points and previous use in UTs for very short term measurements) due to their high thermal neutron capture cross sections, as both are known to be prone to decalibration due to transmutation. Molybdenum and niobium have high melting points and low thermal neutron capture cross sections. Variations of these materials, KW-molybdenum (molybdenum doped with small amounts of potassium, silicon, and tungsten) and niobium-1% zirconium were selected for initial testing. Prior experience with these materials indicates that they retain ductility better than pure metals after heating. For lower temperature tests (less than 1000 ^{o}C) in liquid metal or liquid sodium bonded metallic fuels, stainless steel and Inconel were selected for cost, corrosion resistance, and ease of fabrication.

The materials selected for initial evaluations are listed in Table II.

Table I. Candidate sensor materials.

Material	Melting Temperature (^{o}C)	Thermal Neutron Capture Cross Section (barns)[1]	Possible Irradiation Test Application
302 Stainless Steel	1510	3.02	Liquid Metal Bonded Metallic Fuel (T<1000 ^{o}C)
304 Stainless Steel	1510	3.03	
Inconel 606	1400	4.35	
Molybdenum	2620	2.51	Inert Gas Filled Ceramic Fuel (T≤2600 ^{o}C)
KW-Molybdenum	2620	2.51	
Nb-1% Zr	2470	1.14	

2.2 Sample Fabrication

Test samples consist of a wire waveguide, with a short section isolated by an acoustic reflector (similar to a single sensor UT of the type described in Figure 1). A magnetostrictive wire segment was welded to the end of each waveguide and used, in conjunction with a coil, to generate and sense the acoustic pulses and reflections.

Reflections are produced when an incident acoustic pulse encounters an acoustic discontinuity (an acoustic impedance mismatch). The characteristic acoustic impedance of a waveguide can be defined as:

$$Z = \frac{\rho \cdot c}{A} \qquad (2)$$

Where:

[1] Thermal neutron capture cross sections are calculated as a weighted average of the cross sections of major constituent elements [4].

Z is the impedance,

ρ is the material density,

c is the acoustic velocity (a function of density and stiffness),

and A is the cross-sectional area.

A sudden change in any of these parameters will produce a reflection from an incident acoustic pulse. The simplest method for creating a reflector in a wire waveguide is by introducing a sudden change in the cross-section. Three types of reflectors were required due to differences in the sample wires. Large diameter (1.58 mm) samples could be easily machined with a circumferential notch. Smaller diameter (0.254 mm) samples became unacceptably fragile after machining. For these samples, a dilation was formed by laser welding a short section of the sample wire around the circumference of the sample. The niobium alloy sample was observed to become extremely brittle due to localized heating and minor surface oxidation from laser welding. Hence, reflectors were created on the niobium sample by simply adding a sharp bend to the wire. The three methods used to form reflectors (notch, dilation, and double bend) for this test are shown in Figure 2.

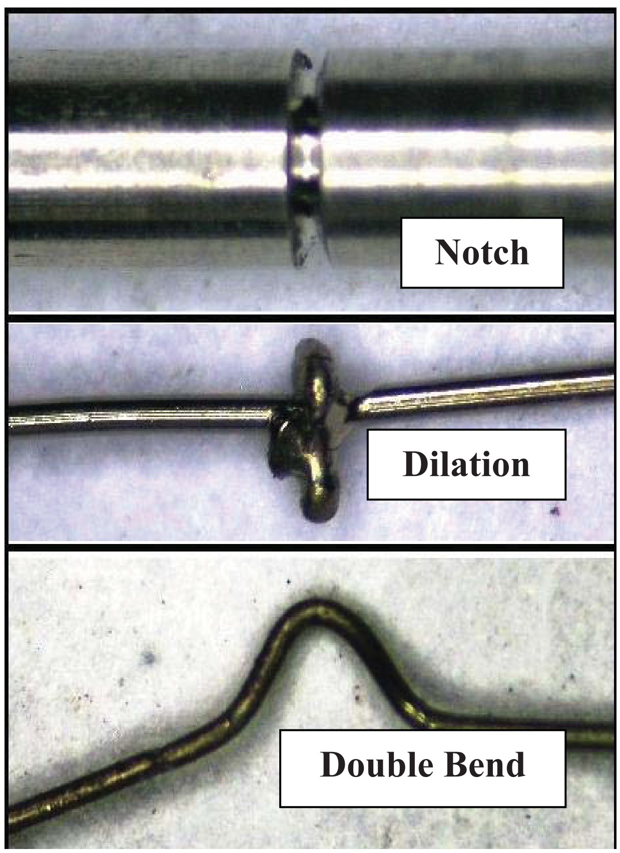

Figure 2. Reflector geometries used in velocity characterization test.

The samples included in this test are listed in Table II.

Table II. Tested samples.

Material	Sample Diameter (mm)	Sensor Length (mm)	Reflector Type
302 Stainless Steel	0.254	79.4	Dilation
304 Stainless Steel	1.58	76.7	Notch
Molybdenum	1.58	77.0	Notch
KW-Molybdenum	0.254	77.0	Dilation
Inconel 606	0.254	76.7	Dilation
Nb-1% Zr	0.254	79.5	Double Bend

2.3 Test Setup

Test specimens were isolated from each other in alumina tubes, which were installed in a tube furnace equipped with an argon purge gas system (see Figure 3 which shows a single installed sample, a total of six were included in the test). Signals were generated using a commercial pulser/receiver system and coils fabricated in-house. Data were monitored and recorded using a high speed digital oscilloscope. Temperatures were monitored using a National Institute of Standards and Technology (NIST) traceable Type-S thermocouple. Data were collected in 100 ^{o}C increments from room temperature to 1300 ^{o}C.

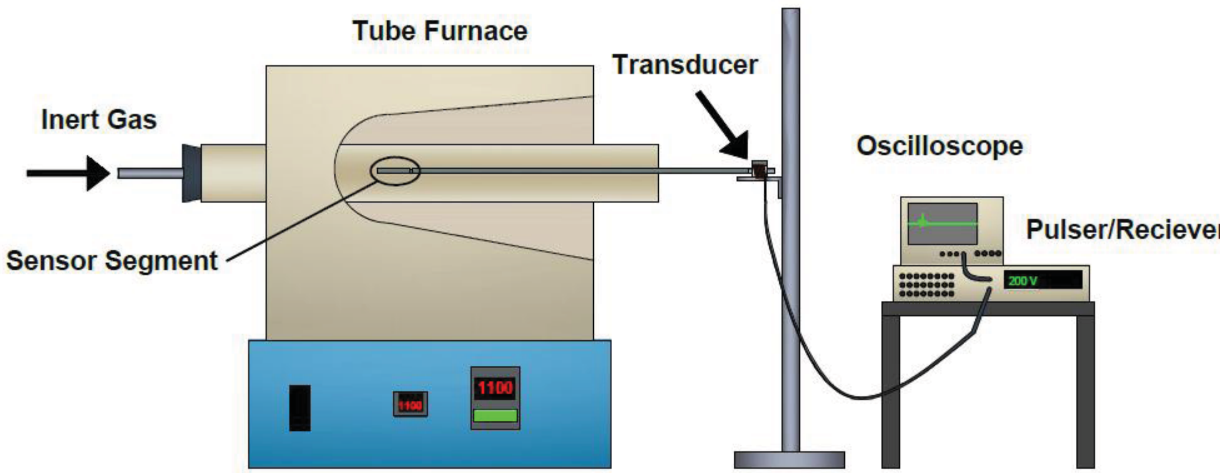

Figure 3. Schematic diagram of test setup.

2.4 Signal Processing

A signal processing method described by Roberts, et al. [5], was implemented in this test. This method consists of cross-correlating the time series data to a known signal (typically either the input signal or the expected reflection), then squaring and low pass filtering the correlated data. This method greatly increases the signal-to-noise ratio (SNR) and simplifies identification of reflections, even in the presence of significant noise. This makes the technique especially useful when reflected wavelets are noisy and when reflection shape varies between test specimens. The input signal used in this test was a simple square pulse, and could not be used in the cross-correlation. The reflection signals were observed to vary from sample to sample, due to differences in material, reflector geometry, quality of welds between samples and the magnetostrictive wire used to generate signals, etc. Therefore, a computer program (using commercially available mathematics software) was developed that allows the graphical selection of a feature, ideally a relatively clear reflection, of the recorded data to use for cross-correlation. Conditioning of the data is then done automatically. Maxima of the filtered data can then be graphically selected, and the velocity is calculated from the delay time between these maxima. A graphical example of this process is shown in Figure 4.

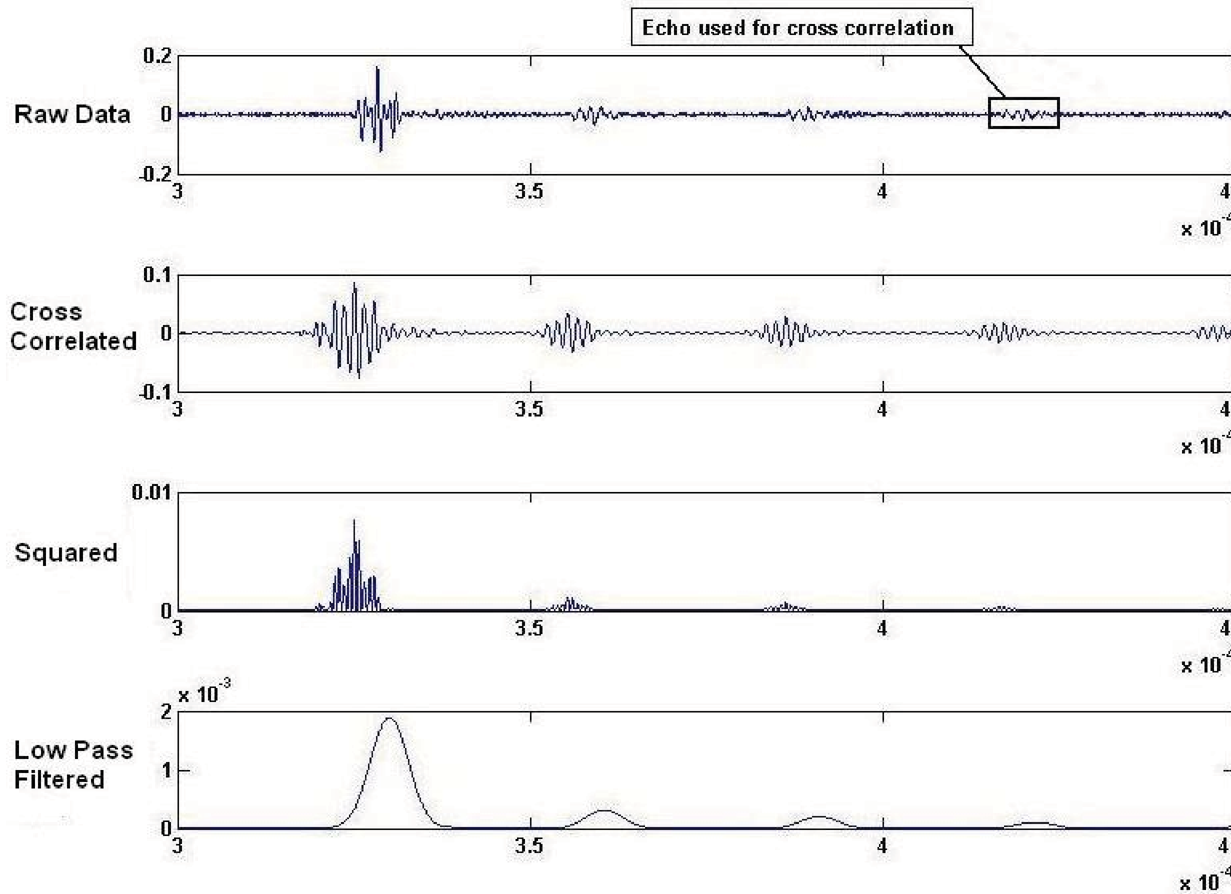

Figure 4. Graphical representation of data analysis process.

Acoustic velocity was calculated from the delay time between the maxima in the filtered data and the known length of the sensor segment (after correcting for thermal expansion) using the following equation:

$$c = \frac{2(l + \Delta l)}{\Delta t} \tag{3}$$

Where:

c is the acoustic velocity,

l is the initial sensor length at room temperature,

Δl is the change in length due to thermal expansion,

Δt is the delay time between maxima.

3 TEST RESULTS

This section details results of the test. Results for measured acoustic velocity for the test samples are compared to values calculated from reference values of Young's modulus and bulk density. Anomalous results observed for Inconel and niobium samples are also briefly discussed.

3.1 Acoustic Velocity of Test Materials

For comparison to velocity values derived in the current work, the acoustic velocity was calculated from reference values of density and elastic modulus using Equation (1). Differences between derived acoustic velocities and those calculated from reference data are attributed to differences in heat treatment, cold work (all tested samples were in an "as drawn" condition), and minor variations in composition. All of these factors can affect the elastic modulus of the material and, therefore, the acoustic velocity. As the history of each test specimen and that of the reference materials is not well known, a direct comparison is impossible.

3.1.1 Stainless Steel

Figure 5 compares measured acoustic velocity of 302 and 304 series stainless steel samples to values calculated from reference data [6,7]. Although the velocity values of the tested samples are somewhat higher (about 5% for temperatures below 800 ^{o}C) than values found in the literature, the trend is consistent and appropriate for UT applications. High temperature attenuation increased signal noise and eventual loss of signal for temperatures above 1100 ^{o}C for the 0.254 mm diameter sample and above 1200 ^{o}C for the 1.58 mm diameter sample. It was also observed that the increased signal noise remained after cooling the sample.

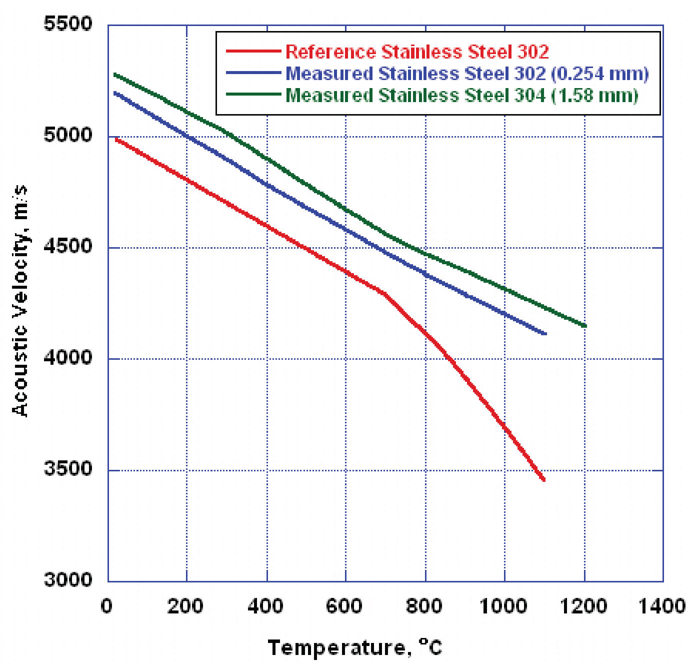

Figure 5. Comparison of measured acoustic velocity of stainless steel to calculated reference values.

3.1.2 Molybdenum

Figure 6 compares measured acoustic velocities for pure molybdenum and KW-molybdenum samples to values calculated from reference data [6,8]. The temperature response of the molybdenum samples is close to values found in the literature, except at the highest and lowest test temperatures. Signals for molybdenum samples did not attenuate significantly over the temperature range of this test.

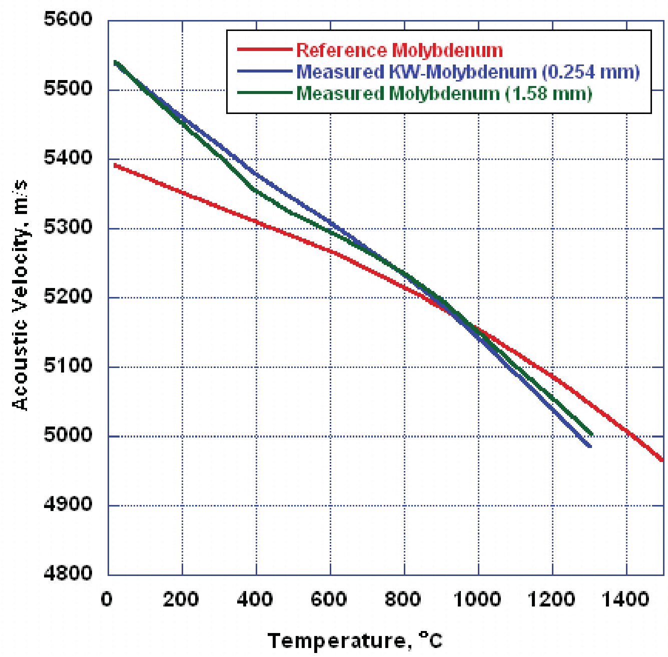

Figure 6. Comparison of measured acoustic velocity of molybdenum to calculated reference values.

3.1.3 Inconel

Figure 7 shows the acoustic velocity of Inconel 606 to values calculated from reference data [6,7]. Although the velocity values of the tested samples are somewhat higher (better than 5% for temperatures up to 1100 °C) than the reference, the trend is consistent and appropriate for use as a UT.

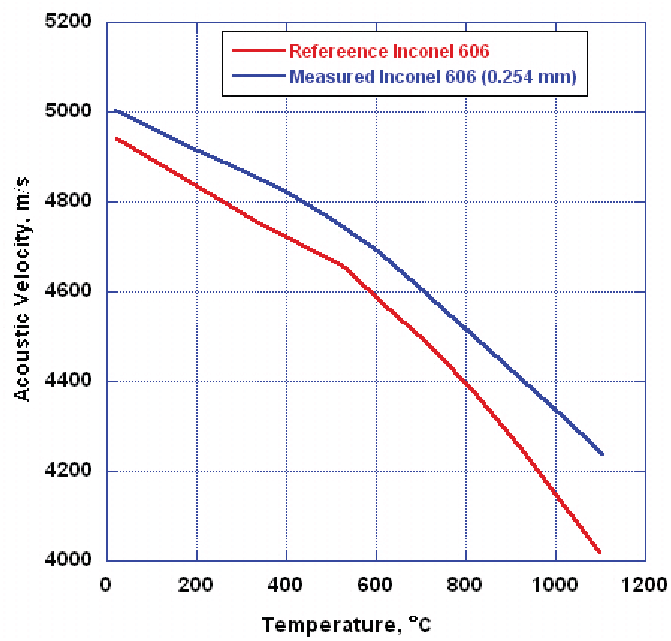

Figure 7. Comparison of measured acoustic velocity of Inconel to calculated reference values.

Sticking, a form of contact bonding between the sensor and its surroundings that can interfere with and obscure acoustic signals, occurred for Inconel 606 at temperatures above 1000 °C. Sticking was only observed at temperatures above 1800 °C in previous research [9,10,11]. This sticking could have been due to contact bonding between the sample and the alumina tube, but it also may have been due to binding between the sample and the tube caused by thermal expansion. An example of the effect of sticking is shown in Figure 10 for an Inconel sample tested at temperatures between 1000 °C and 1300 °C. Sticking appears to begin at 1100 °C and has completely obscured the reflector signal at 1200 °C.

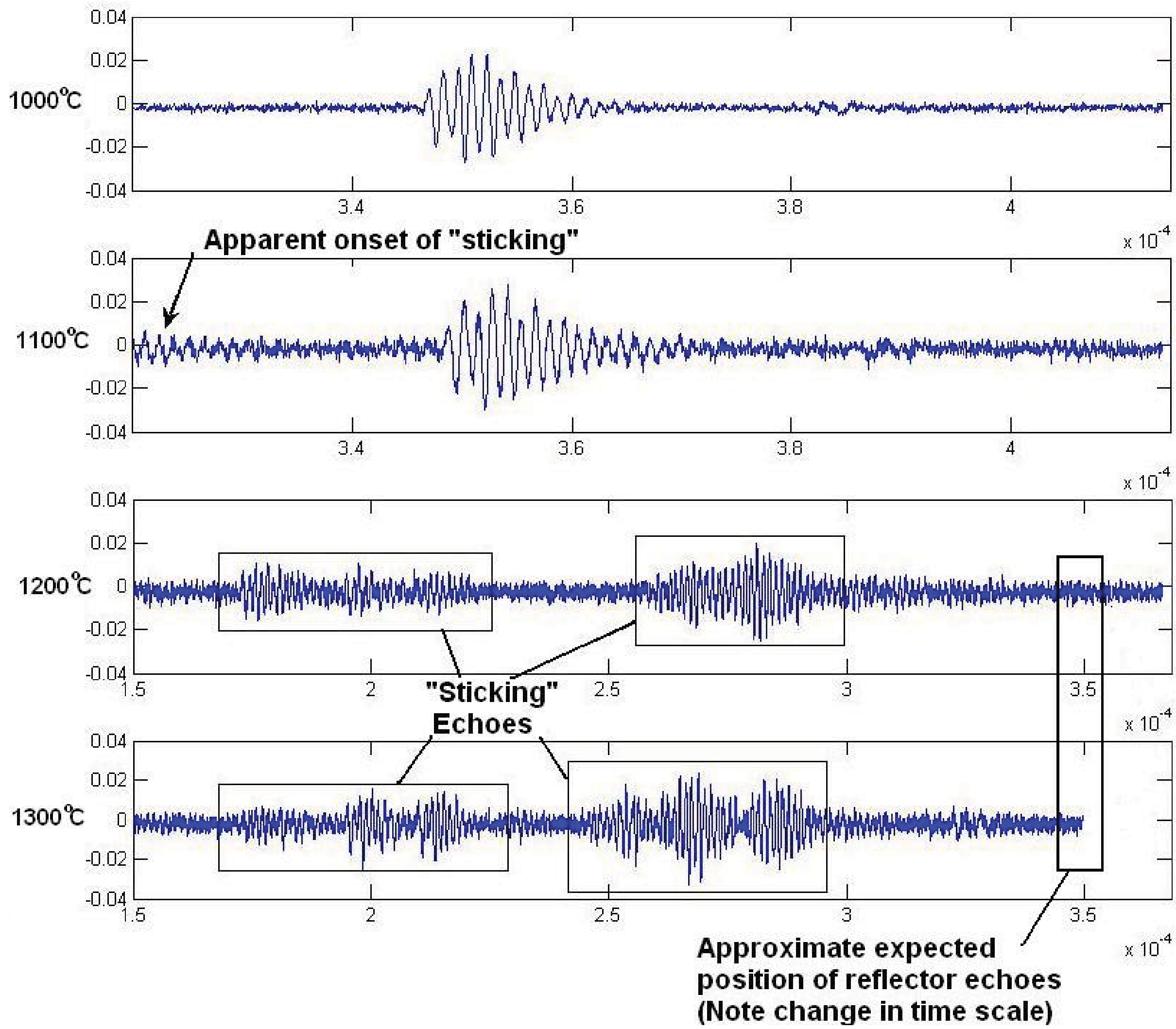

Figure 8. Progression of "sticking" observed for Inconel sample.

3.1.4 Niobium-1% Zirconium

The temperature response of the niobium-1% zirconium sample differed from values calculated from reference data, although the velocity values are close at room temperature. Reference data for density [12] and elastic modulus [13] predict a decrease in acoustic velocity with increasing temperature, as for the other tested samples. However, recorded data show an increase in velocity with temperature, as seen in Figure 8.

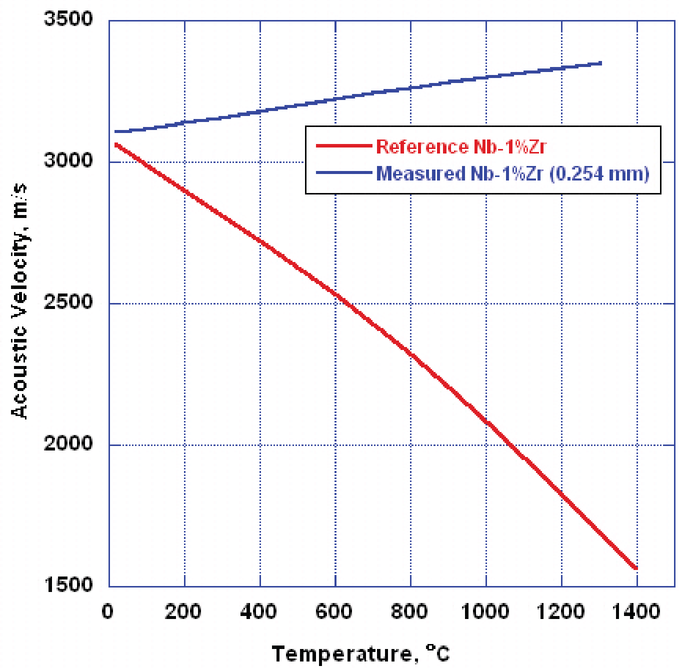

Figure 9. Comparison of measured acoustic velocity of Nb-1%Zr to calculated reference values.

Difficulties in fabricating the niobium alloy sample could explain this discrepancy. For example, welding the niobium to the magnetostrictive stub with a laser welder was observed to significantly embrittle the wire. To overcome this, a small amount of nickel wire was melted onto the weld in order to strengthen the joint, causing a slight bulge. The embrittlement also made the "dilation" type reflector problematic and led to the use of the double bend reflector. Ringing in the magnetostrictive stub, due to the diameter increase, was observed to cause the acoustic pulse to spread. The double bend reflector, as it was not a sharp acoustic discontinuity, could have also increased measurement uncertainties. The "spread out" nature of the signals can be seen in Figure 9, which shows two reflection signals used to calculate acoustic velocity. Although the non-ideal signals could lead to increased error in the calculation of acoustic velocity, a comparison of signals recorded at different temperatures does seem to verify the increase in velocity. Lines of equal slope were used to connect an easily identifiable feature common to both reflections starting at 20 ^{o}C. The line perfectly connects this feature in the leading reflection (at around 5×10^{-4} s). However a slight adjustment to the slope was required to fit the trailing reflection. This indicates a reduction in delay time with increasing temperature and, therefore, an increase in acoustic velocity.

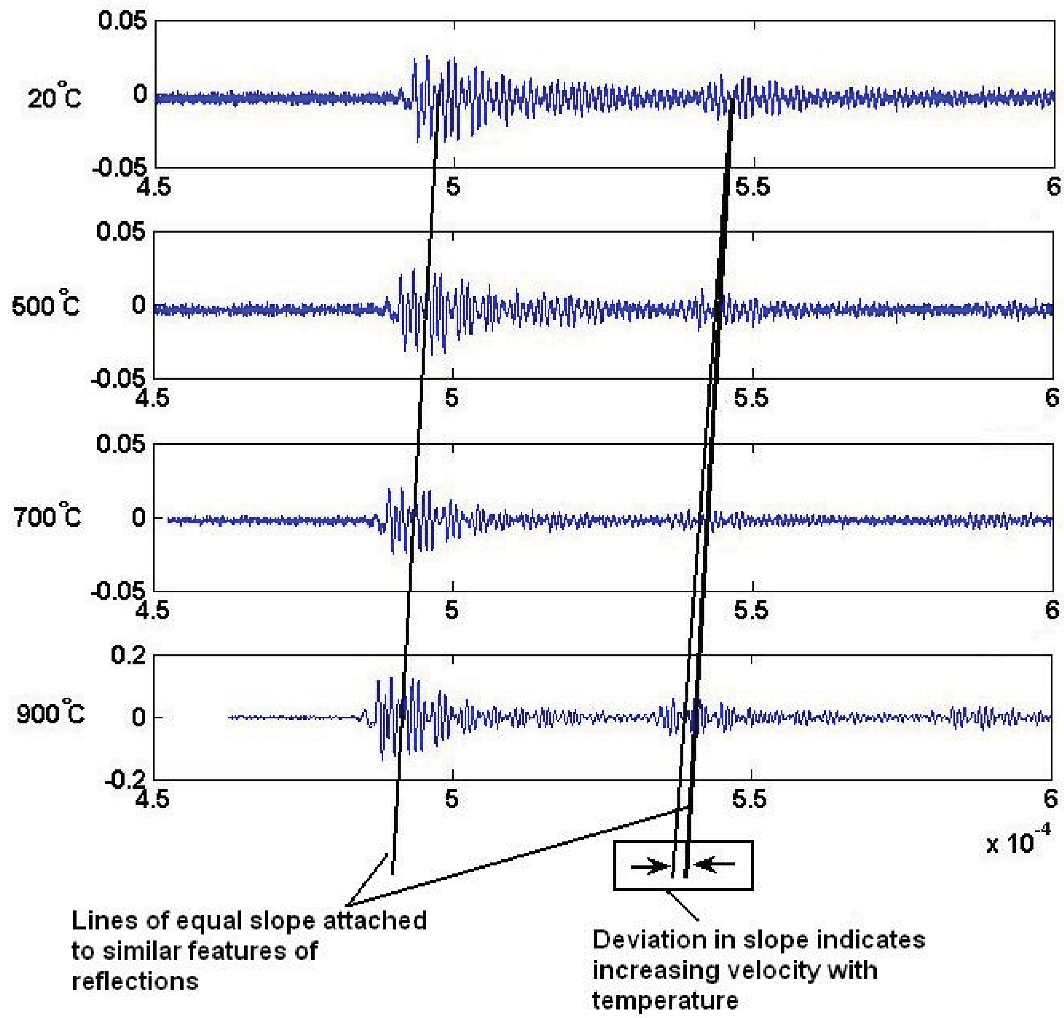

Figure 10. Time series data for Nb-1%Zr sample showing increase in acoustic velocity with temperature.

4 SUMMARY AND FUTURE WORK

Design and testing of an ultrasonic thermometry system for in-pile temperature monitoring has been initiated. Several candidate materials appropriate for use in inert gas filled ceramic fuel tests and liquid sodium filled metallic fuel tests have been identified. The temperature response of the acoustic velocity of these materials was evaluated for temperatures up to 1300 °C.

Results indicate that both tested stainless steel samples and the Inconel sample produce signals of good quality over the temperature range of interest for these materials (up to 1000 °C). Results indicate that both tested molybdenum based samples produce signals of good quality over the tested temperature range (up to 1300 °C). Due to the difficulty in fabricating samples, the low durability of the welds, and poor temperature sensitivity, niobium based alloys will not be considered in future work.

A signal processing technique has also been identified and adapted for supporting such testing. This technique has been demonstrated to improve signal to noise ratio and to be insensitive to sample-to-sample changes in waveform shape.

Future work will include testing of molybdenum samples at higher temperatures (up to at least 2600 °C) and development of a method to eliminate sticking. Further development of signal processing capabilities will also be required to overcome the problem of overlapping reflections. Finally a prototype system will be developed and tested to evaluate the potential accuracy and resolution possible with a UT system.

5 ACKNOWLEDGMENTS

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