

# Results of NDE Technique Evaluation of Clad Hydrides

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**Fuel Cycle Research & Development**

***Prepared for  
U.S. Department of Energy  
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Dennis C. Kunerth  
National Laboratory  
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## SUMMARY

This report fulfills the M4 milestone, M4FT-14IN0805023, Results of NDE Technique Evaluation of Clad Hydrides, under Work Package Number FT-14IN080502.

During service, zirconium alloy fuel cladding will degrade via corrosion/oxidation. Hydrogen, a byproduct of the oxidation process, will be absorbed into the cladding and eventually form hydrides due to low hydrogen solubility limits. The hydride phase is detrimental to the mechanical properties of the cladding and therefore it is important to be able to detect and characterize the presence of this constituent within the cladding. Presently, hydrides are evaluated using destructive examination. If nondestructive evaluation techniques can be used to detect and characterize the hydrides, the potential exists to significantly increase test sample coverage while reducing evaluation time and cost. To demonstrate the viability this approach, an initial evaluation of eddy current and ultrasonic techniques were performed to demonstrate the basic ability to these techniques to detect hydrides or their effects on the microstructure.

Conventional continuous wave eddy current techniques were applied to zirconium based cladding test samples thermally processed with hydrogen gas to promote the absorption of hydrogen and subsequent formation of hydrides. The results of the evaluation demonstrate that eddy current inspection approaches have the potential to detect both the physical damage induced by hydrides, e.g. blisters and cracking, as well as the combined effects of absorbed hydrogen and hydride precipitates on the electrical properties of the zirconium alloy. Similarly, measurements of ultrasonic wave velocities indicate changes in the elastic properties resulting from the combined effects of absorbed hydrogen and hydride precipitates as well as changes in geometry in regions of severe degradation. However, for both approaches, the signal responses intended to make the desired measurement incorporate a number of contributing parameters. These contributing factors need to be recognized and a means to control them or separate their contributions will be required to obtain the desired information.

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# Results of NDE Technique Evaluation of Clad Hydrides

## 1. INTRODUCTION

During service, zirconium alloy fuel cladding will degrade via corrosion/oxidation. As part of the oxidation process, a byproduct, hydrogen, will be absorbed into the cladding and eventually form hydrides due to low hydrogen solubility limits within the metal alloy combined with the service/post service thermal conditions. The hydride phase is detrimental to the mechanical properties of the cladding and therefore it is important to be able to detect and characterize the presence of this constituent within the cladding. Of primary interest are the amount and orientation of the hydride precipitates. Presently, hydrides are characterized via destructive examination. This is time consuming and costly. An alternative is to use nondestructive evaluation technologies to detect and characterize the hydrides. If viable, this approach will significantly increase test sample coverage while reducing evaluation time and cost.

The activities presented in this document summarize the initial effort to examine the viability of using eddy currents and ultrasonic approaches to detect and characterize zirconium hydride in fuel cladding. To be discussed is the development of test samples and the initial measurements using an array eddy current system and laser ultrasonics to detect hydrides or their effects on the microstructure.

### 1.1 Test Sample Preparation

A thermal process was used to develop hydride test samples in which the test samples of interest are placed in a tube furnace containing a flowing cover gas of pure hydrogen. The temperature is then elevated and held at a constant temperature or cycled with a narrow temperature range near the upper set point. The specific zirconium alloy cladding test samples on which the eddy current and ultrasonic measurements are presented in this document are pictured in Fig. 1. These samples were raised to a temperature of 470° C at 10° C/min immediately followed by a cool down at 10° C/min. The hydrogen cover gas was changed to nitrogen during cool down. Due to the nature of the processing, an axial hydrogen/hydride gradient is formed in the test samples. As observed in Fig. 1 the test samples are severely degraded towards the inflow of the hydrogen gas. The severely degraded sections were removed prior to making measurements. Fig. 2 shows the condition of the cladding test samples used for measurements.

### 1.2 Eddy Current Inspection

Eddy current techniques have been used to evaluate nuclear fuel and indirectly determine loss of wall thickness and hydrogen concentration via measurement of the oxide thickness layer formed during oxidation.[1-3] For this evaluation, a commercial array eddy current instrument and probe was used to inspect hydrided zirconium alloy cladding test samples. The array probe was designed to inspect 11.2 mm diameter cladding using a circumferential array of 30 transmit-receive coils and 4 absolute located 90° apart, see Fig. 3. The transmit-coils are intended for defect detection while the absolute coils are intended for oxide thickness measurements. However, due to the nature of the absolute coils, they sense not only the liftoff used to determine oxide thickness but at lower test frequencies, the changes in clad material electrical conductivity due to hydrogen and hydride content. Fig. 4 contains the transverse C-Scan image from the 30 coil transmit-receive array of a short section of hydrided cladding. The transmit-receive coil

arrangement allows transverse and axial C-Scan data to be collected during a single scan. The transverse coils are most sensitive to defects with a transverse orientation while the axial coils are most sensitive to defects with an axial orientation. As demonstrated, this type of eddy current coil arrangement is well suited to detect the physical defects in the tubes as well as pickup anomalous regions due to severe hydriding, fretting damage, or other material conditions that significantly affect electrical conductivity. Fig. 5 contains the signal traces for the absolute coils. It can be observed that there is a gradual increase in signal strength along the axis of the test sample. This is in response to the gradient in hydrogen and hydride concentration generated by the thermal hydriding treatment. This indicates that eddy current inspection technologies can be used for defect detection such as blisters, pitting, and crack as well as provide information about the changes in electrical conductivity of the cladding material due to absorbed hydrogen and hydride formation. Eddy current inspection technologies also have the potential to evaluate the orientation of the hydride phase, e.g. circumferential versus radial.[4, 5] This will require appropriate coil configurations to permit the determination of anisotropic electrical conductivity.

However, note that there are various parameters that contribute to the electrical conductivity. Test material phase, temperature, grain size, stress, hydride content and orientation, and hydrogen content all contribute. If a basic understanding of the different parameters can be obtained, a quantitative estimate of a single contribution can be made. To do this, additional information may be required via implementation of complementary inspection technologies.

### 1.3 Ultrasonic Inspection

Ultrasonic inspection is dependent on the elastic properties of the test material as well as geometry. The presence of absorbed hydrogen and hydride phases will alter the mechanical properties as well as affect the attenuation and scatter of a propagating ultrasonic wave.[6] Besides the results presented here, other researchers have investigated the potential of ultrasonics to detect and characterize hydrides in a zirconium based clad material.[7] Laser based narrow band surface acoustic wave measurements were made on hydrided cladding test samples. The laser ultrasound methods are well established in generating and analyzing surface acoustic waves in materials of the thickness scales comparable to cladding. Laser ultrasonics entails thermoelastic generation of ultrasound by irradiating a specimen with a high intensity laser pulse. Surface waves are confined to propagate along the sample surface and have propagation characteristics that are related to near surface mechanical properties. Lower test frequencies will generate longer wavelengths that penetrate deeper in the test piece. For this measurement, increasing the grid spacing will increase the test wavelength. Fig. 6 provides a description of the narrow band acoustic surface wave measurement, typical response, and experimental setup. Provided in Fig. 7 are the measured results for a hydrided test sample and reference. Although small, there are velocity differences between the two samples. The obvious differences in velocity measured using the two different gratings are the result of dispersion (different wave velocities at different frequencies/wavelengths) and measurement error. Another laser based measurement using a pulsed point source (2.5 mm spot size) and pickup (8 mm separation) generates broadband plate waves. The results of this measurement are provided in Fig. 8. As with the narrow band ultrasonic surface wave measurements, there are differences in the responses. However, the degradation of the inner diameter of the hydrided tube may account for the difference measured by the plate wave. The dispersion in the broad band signals is most noticeable at the lower frequencies (received later in time). The velocity measurements performed are only one type of ultrasonic approach to detecting and characterizing hydrides. Other attributes of the propagating ultrasonic wave such as attenuation or scatter can be added to refine the inspection process.

## 2. CONCLUSIONS

The measurements performed verify the ability of the two inspection approaches to detect physical and microstructural changes within the test samples but do not demonstrate the ability to separate the contributing factors to the signals thereby allowing the desired information to be extracted. Note that implementation of a single technique is typically based on a set of assumptions regarding various test parameters and that it may be useful to combine the results of complementary techniques to fully understand the measurements obtained.

Additional work will be required to refine the inspection approaches and signal processing steps to obtain the desired measurement. Vital to this will be the development of appropriate test samples having known, typical hydride concentrations and orientations. The test samples used to date were not well controlled with respect to hydride concentration but did serve the purpose for this initial evaluation. The basic steps that need to be performed in order to implement nondestructive evaluation approaches to characterizing hydride in fuel cladding are as follows.

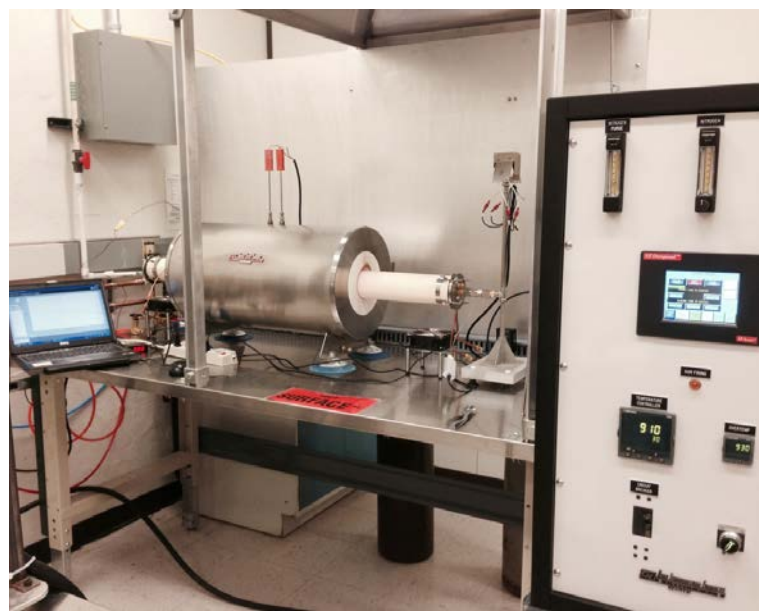
- 1) Identify environmental and test parameters required to be addressed during inspection, e.g. pool side or hot cell environment, radiation levels, temperature, test sample geometries, etc.
- 2) Develop and characterize a set of cladding test samples typical of the hydride concentrations and orientations of interest. Included will be cladding alloy and geometry.
- 3) Identify and test, using the sample set developed in step 1, the eddy current, ultrasonic, or alternative nondestructive evaluation technique(s) deemed to be appropriate for the stated test parameters and the desired measurements. This includes the development and testing of hardware as well as signal processing approaches to correlate the collected data with the known test sample material conditions. This is an iterative process to refine and verify the validity of the inspection process as well as characterize its capabilities/limitations.
- 4) Formalize the inspection's technical basis, measurement technique, required hardware, signal processing algorithms, and methods of calibration for use by industry.

## 3. REFERENCES

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#### 4. FIGURES



Laboratory Hydriding Furnace

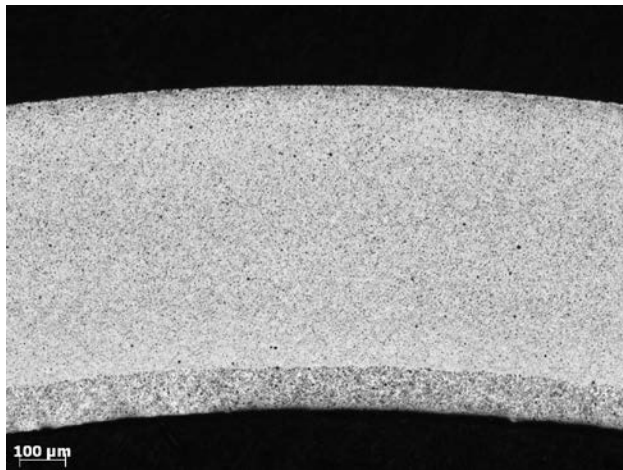


Before Processing

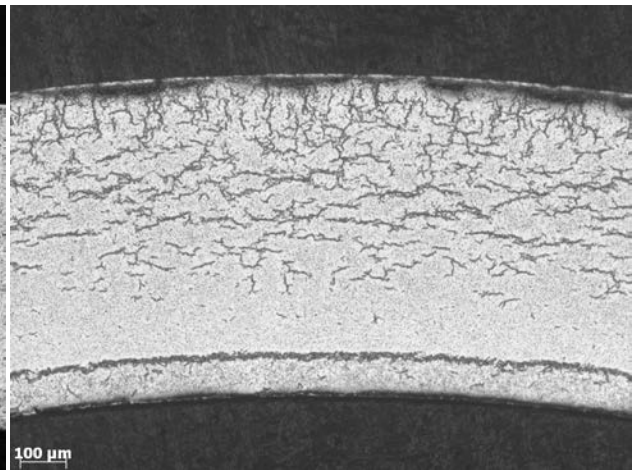


After Processing

Figure 1. Laboratory system used to hydride test samples. Right: Test samples before and after processing.



Reference Cladding



Hydrided Cladding

Figure 2. Cladding test samples used during evaluation of the nondestructive evaluation approaches.  
Bottom: Microstructure of unprocessed reference cladding and a hydrided test sample.

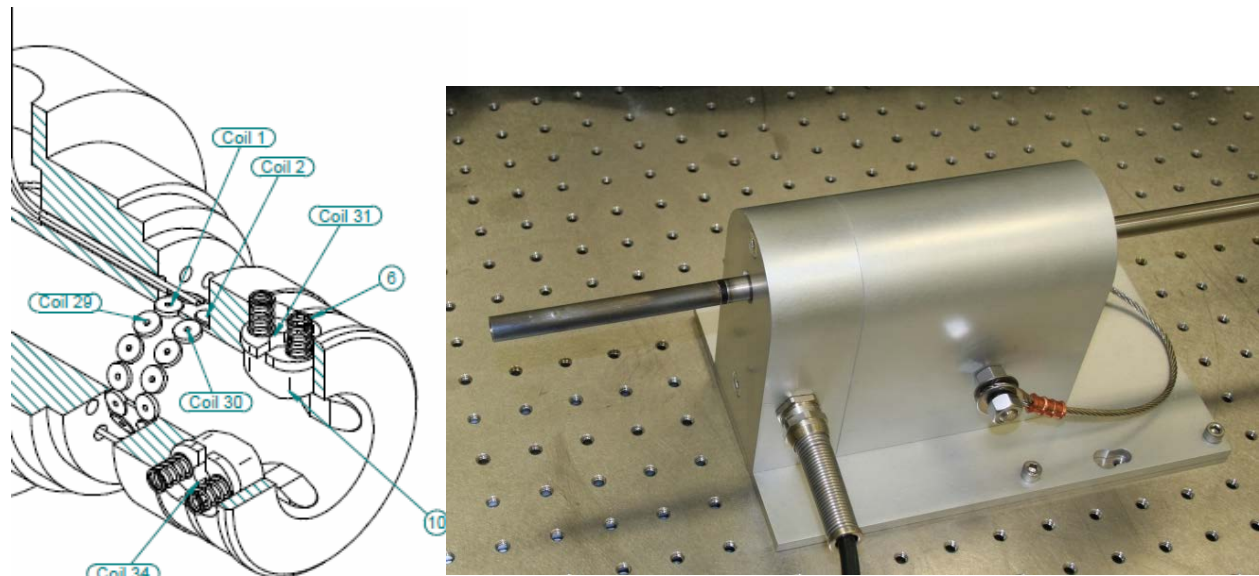


Figure 3. Basic coil arrangement and image the array eddy current probe used to scan the cladding test samples.

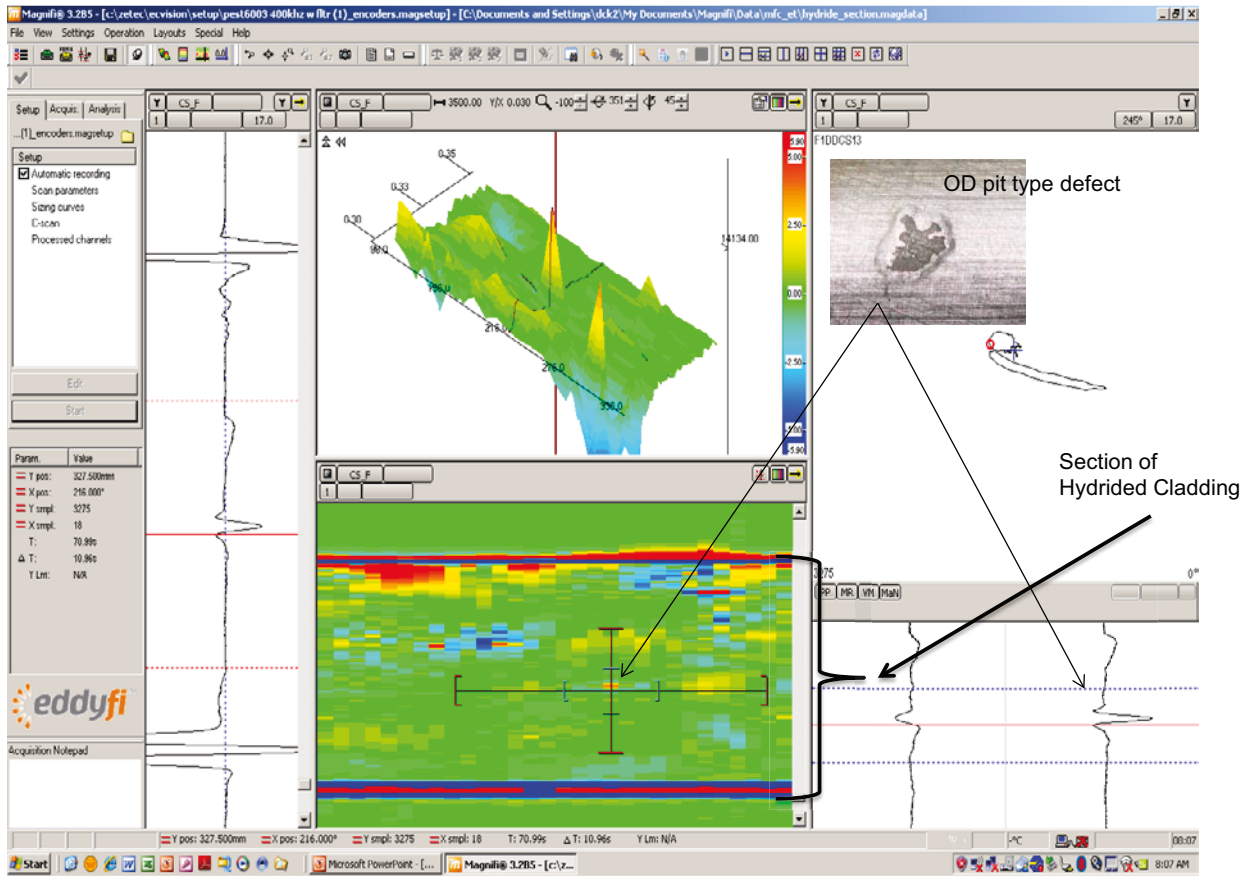


Figure 4. Transverse C-Scan image of a hydrided test sample. The central colored coded images are the 2D and isometric C-Scan images. On the right are the impedance plane response with the individual 0° and 90° impedance component traces provided below. The left hand window is the expanded view of the 90° component trace. The cursor in the C-Scan is centered over the response of the outside diameter defect pictured in the impedance plane window. Besides the isolated surface defect highlighted, various other flaw indications are present in the C-Scan image.



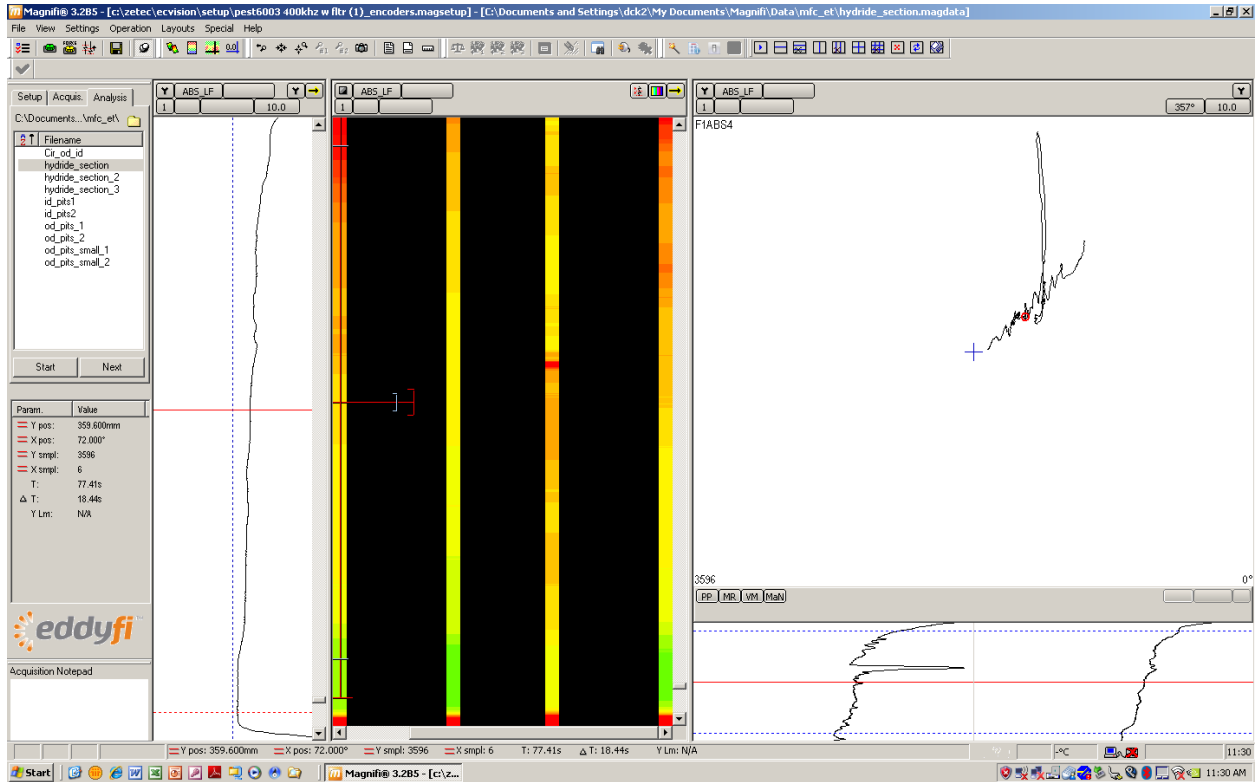
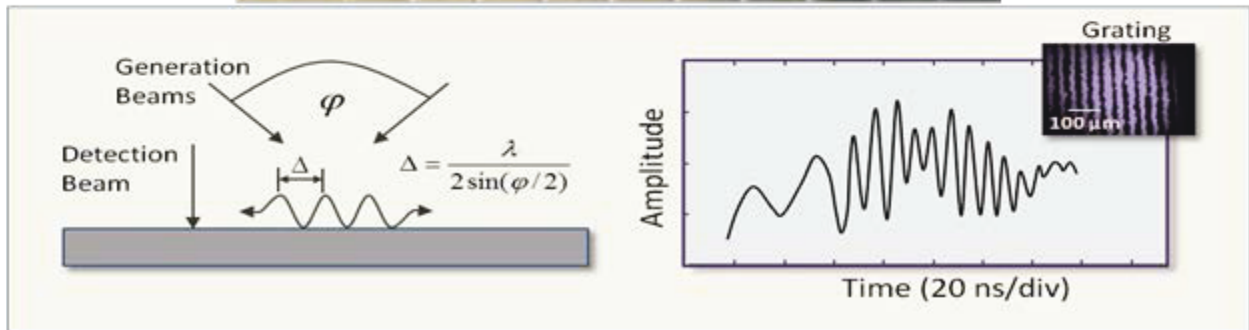
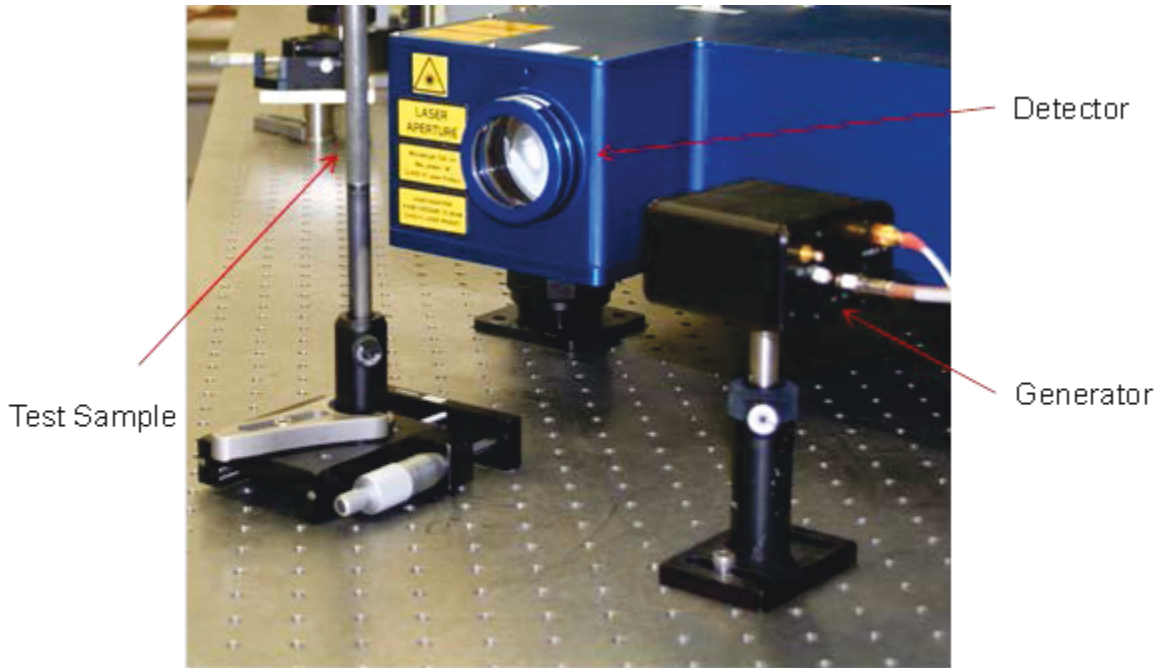
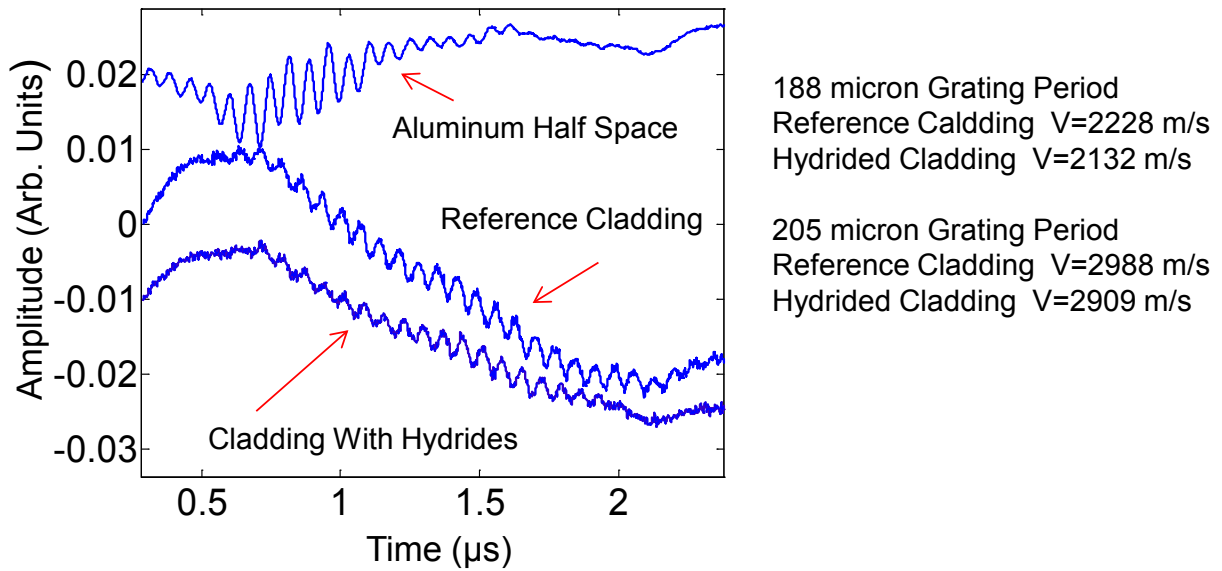


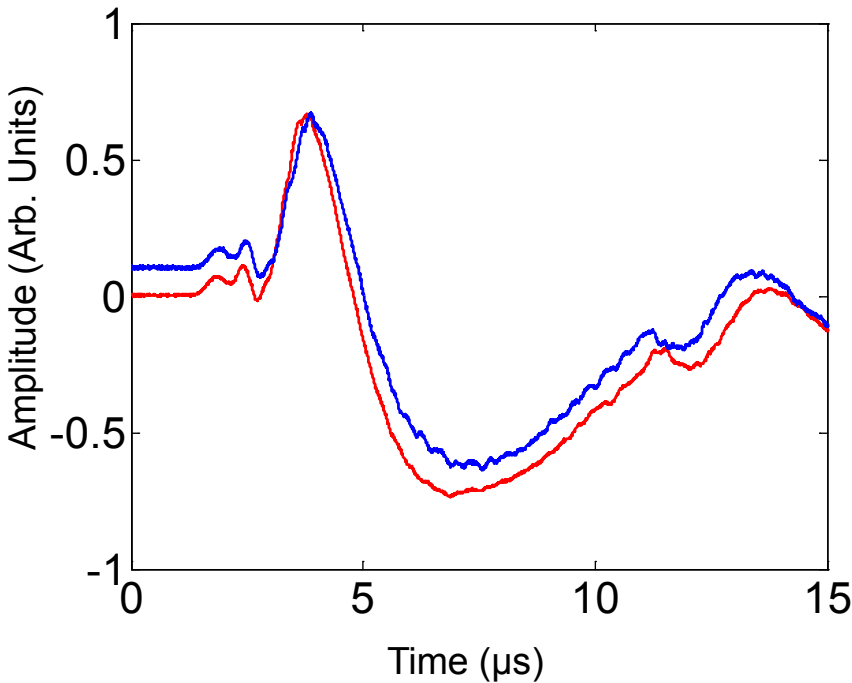
Figure 5. Scan traces for the four absolute coils in the array eddy current probe. The cursor is centered over the 0° coil trace exemplifying the gradual change in response along the scan direction. The gradual change in signal amplitude is indicative of a gradient in electrical conductivity.



**Figure 6.** Experimental setup for generating narrow band surface acoustic waves. Lower Right: Grating and typical data for laser generated narrowband surface acoustic wave.



**Figure 7.** Narrow band acoustic surface wave results for reference and hydrided cladding samples. Right: Differences in measured acoustic velocity for two different grating periods. The differences in acoustic velocity between the two different gratings are a result of dispersion and measurement error.



**Figure 8.** Broad band ultrasonic plate wave results for reference and hydrided cladding samples.