UDRI

UNIVERSITY
of DAYTON

RESEARCH
INSTITUTE

UDR-TR-2004-00018

Report

MONITORING OF REFRACTORY WALL RECESSION USING HIGH-TEMPERATURE IMPACT-ECHO INSTRUMENTATION

.....
March 2004

James Sebastian

Monitoring of Refractory Wall Recession Using High-Temperature Impact-Echo Instrumentation

Final Report

Contract: DE_FG36-02GO12051

March 2004

Prepared for:

U.S. Department of Energy
Golden Field Office
Golden, CO 80401

Prepared by:

James Sebastian, P.E.
STRUCTURAL INTEGRITY DIVISION
300 College Park
Dayton, OH 45469-0120

Section 1

Introduction

Regression of refractory linings of furnaces occurs due to a variety of mechanisms. The specific mechanism selected for investigation during this program is the regression of refractories which are in direct contact with a liquid corrodant. Examples include the melting of glass, the production of pig iron and steel, and the melting of aluminum. The rates of regression to a wall thickness which requires reline or extensive reconstruction vary widely, from less than a year to over ten years depending on the specific service environment.

This program investigated the feasibility of measuring refractory wall thickness with an impact-echo method while at operating temperature (wall temperatures exceeding 500°C). The impact-echo method uses the impact of a small sphere with the surface of the test object to send a stress wave into the object. In a plate-like structure, the stress wave reflects back to the front surface, reverberating in the structure and causing a periodic surface displacement whose frequency is inversely proportional to the thickness of the test object. Impact-echo testing was chosen because it requires access to only one side of the test object and could be performed during the operation of a refractory structure. Commercially-available impact-echo instrumentation is available for room temperature use for a variety of tests on concrete. The enabling technology for this work was to use a high-temperature piezoelectric material, aluminum nitride, as the receiving sensor for the stress waves, allowing its use on refractories during furnace operation.

The University of Dayton wishes to thank the Ceramic Engineering Department at the University of Missouri-Rolla for advice and support during the project. Dr. Mariano Velez, Dr. Bill Headrick, and Thomas Burns provided valuable information, test samples, and the use of facilities for collaborative efforts.

Section 2

Background

2.1 Aluminum Nitride (AlN) Sensor Technology

Commonly-used piezoceramic materials, such as lead zirconate titanate (PZT), can be used in sensors and transducers up to approximately 250°C for brief periods. Special cuts of quartz can be used to approximately 350°C. Lithium Niobate (LiNbO_3), a ferroelectric material, can be made to work at high temperatures, but only under very carefully-controlled conditions due to its fragility and need for a controlled oxygen environment. PZT is made piezoelectric by an elevated temperature poling process; as the poling temperature is approached, the sensor becomes permanently depoled and is no longer piezoelectric. Although quartz is naturally piezoelectric and does not require poling, it undergoes a change in crystal structure at its Curie temperature, which removes its piezoelectric properties until it is cooled. In contrast, AlN is also naturally piezoelectric, but has no known Curie temperature, which allows it to remain piezoelectric at temperatures exceeding 1000°C.

The piezoelectric properties of aluminum nitride (AlN) have been known for over thirty years. Very thin AlN films ($<1\text{ }\mu\text{m}$) are used commercially in filters for radio frequency electronics. In general, these films are too thin for use as a bulk piezoelectric sensor material, and the production of thicker films by the methods used for filter devices, such as sputtering, is cost- and time- prohibitive. Approximately ten years ago, a process was developed which allows the deposition of substantially thicker piezoelectric AlN films, up to 0.15 mm thick. This development made the production of AlN-based sensors and transducers a real possibility.

In 1994-1996, the United States Air Force funded a research program at the University of Dayton to prove the feasibility of using an AlN film as an ultrasonic transducer in a Hot Isostatic Press (HIP). The transducer operated above 900°C at pressures exceeding 150 MPa in an argon environment inside a working HIP vessel. As a result of this success, follow-on efforts were undertaken to further explore uses for AlN films.

The success of this program led to additional research targeted to film improvements, also sponsored by the United States Air Force. This effort took the unreliable film deposition process used in the HIP sensor research and refined it. Film deposition became reliable, with

90% of the films surviving the deposition process. The population of point defects in the films was dramatically reduced. More importantly for ultrasonics, thicker films (up to 150 μm) were produced. These thicker films produce lower frequency energy (below 30 MHz), important for penetration in many materials. Film thickness became a controllable variable.

Parallel efforts evaluated the suitability of the films for a variety of applications and environments. The upper limit of 700°C operation in air was established, with degradation primarily due to substrate oxidation. Operation of the AlN films was demonstrated in molten metals, including solder alloys, zinc, and aluminum. A load cell for high-temperature operation was designed and demonstrated using an AlN ultrasonic transducer. The physical durability of the films was established, with films continuing to operate under a compressive stress exceeding 1 GPa, and surviving without damage.

This research also identified the potential of the AlN films to operate as direct piezoelectric sensors for dynamic force, pressure, and acceleration. These sensors were constructed along the lines of lower-temperature sensors similar to those offered commercially by companies including Kistler, Endevco, and PCB. Sensors were tested up to 400°C in direct force and pressure measurement applications. A related Air Force test program installed sensors in forging dies for the direct measurement of workpiece pressure during the forging process. Two sensors successfully recorded workpiece pressures and die filling of a cylinder of AA6061 as it was forged into a ring shape with a center web. The workpiece was at 530°C for this test.

The research projects described above have proved the viability of the AlN films for use as a piezoelectric sensor material at high temperature. In the current program, AlN films were evaluated for use as impact-echo sensors. This entailed use in the 10kHz to 100 kHz frequency range which had not been previously explored.

2.2 Impact-Echo Testing

Impact-echo testing was developed as a nondestructive testing technique which could be used on concrete and masonry structures in a way similar to pulse-echo ultrasonic testing and can best be described by analogy with ultrasound. Pulse-echo testing uses a piezoelectric transducer to send a high-frequency sound pulse into a material, which interacts with the material and returns to the transducer. It is then converted to an electronic signal, amplified, and evaluated. The problem with using ultrasound on masonry structures is that typical ultrasound

waves used are too high frequency (>100 kHz and generally >500 kHz) for the material, where they are quickly scattered and attenuated. Impact-echo testing solves this problem by using an impact on the surface to create a relatively high-energy stress wave at lower frequencies (<80 kHz). A different transducer design is also employed to receive the reflected stress waves. Originally developed for acoustic-emission work, it uses a conical piezoelectric element with a large backing mass to detect surface displacements.

Impact-echo is used commercially for evaluation of new and in-service masonry structures. Examples include slab and pavement thickness, void location, and delamination of reinforcing steel. In many cases, it has reduced or eliminated the need for expensive and destructive core testing. The simplest application of the method is for thickness testing, the object of this project. In the frequency spectrum of the received signal from the transducer, there is a resonance peak corresponding to the thickness of the material. A proper impactor must be selected to insure sufficient excitation near this frequency; in practice this is done with a series of various-sized steel spheres.

Because impact-echo is a resonance method, knowledge of the speed of sound in the material is required for accurate thickness measurements. The speed of sound varies with composition and density in masonry structures, and is often measured using two transducers a known distance apart on the surface of the structure, aligned with a single point of impact. This will be more complex on the surface of an operating refractory, where the absolute temperature and the temperature gradient through the material also affect the wave speed. Sansalone^[1] provides a complete description of the impact-echo method and history.

Section 3

Initial Testing

Efforts during the initial phase of the project concentrated on gathering preliminary data that would lead to the design of an AlN-based impact-echo transducer. In addition, a commercial impact-echo system was rented in order to gain some experience with the method, insight into its operation, and to test on the refractory material. Impact-echo transducers are similar in construction and function to acoustic emission transducers^[2], and acoustic emission has been used for refractory materials^[3].

The first step toward transducer design was to evaluate piezoelectric film configurations for sensitivity as an impact-echo receiver. The two basic configurations were explored. The simplest configuration was to apply a metal foil to the surface of the test object and to place the AlN film directly on the foil. With the foil forming a first electrode and the substrate attached to the AlN film forming a second electrode, this configuration successfully received stress waves from a test object when the device was sonically coupled to the object by the foil and a compressive force. This test was very encouraging, as the signals could be picked up directly by an oscilloscope without amplification. The next challenge was to reconfigure the device for incorporation in a practical sensor, leading to the second configuration.

This second design inserted a titanium cone between the test object and the aluminum foil, eliminating the need for foil at the surface of the test object, which would simplify use of the device. The conical shape removed the requirement that the test object have a smooth, flat surface, and was modeled on the commercial impact-echo transducer's conical piezoelectric crystal. This design also successfully received signals, with a response similar to that of the commercial transducer, as shown in Figures 1 and 2. The lower amplitude from the AlN transducer is a result of no amplification of the signal; the commercial transducer having built-in amplification. Figure 1 shows simultaneous acquisition from both transducers from a single impact on a ceramic block; the time delay resulted from the different spacing of the two transducers relative to the impact point. The different locations of the transducers also likely caused the signals to look somewhat different, although they have similar frequency content, the most important information extracted from the signal in impact-echo analysis.

Figure 1. Impact-echo signals from ceramic block.

The signals in Figure 2 were acquired separately with the two transducers located in approximately the same location on the building floor, which was used as a concrete slab test object. The impactor used was a 0.25 inch tungsten carbide ball, which imparted more energy than the standard steel ball impactor. The 16 kHz frequency corresponds to a thickness of approximately 5 inches, assuming a 4000m/s wave speed for the concrete.

The above data and similar tests were encouraging for the use of an AlN transducer. However, impact-echo tests on refractory material, such as the fuse-cast alumina-zirconia-silica (AZS) targeted for the high-temperature tests, were less encouraging. These materials have a much higher wave speed than concrete and small particle sizes. The pieces available did not seem to lend themselves to thickness measurements – too many resonant modes were being excited in the blocks. Efforts were begun to acquire plates of test material with higher aspect ratios.

Figure 2. Impact-echo signals from concrete slab floor.

Section 4

AlN Transducer Design

4.1 Aluminum Nitride (AlN) Film Development

The key to a successful high-temperature transducer in this program was functional piezoelectric AlN films. UDRI's deposition system used for previous programs requiring AlN films was used for this work. Previous AlN work usually focused on depositing the AlN on tungsten carbide (WC) cermet substrates, and deposition was achieved on titanium metal at the end of a recent program. Advantages to titanium substrates include improved ultrasonic pulse shapes, facilitate substrate shaping, and increased maximum use temperature of the film-substrate combination.

Initially, problems occurred in producing the films on the titanium substrates for the current project. The films that were produced tended to have poor adhesion and exhibited other undesirable defects such as porosity. Many of these problems were eventually attributed to the titanium alloy being used, and film adhesion and quality rapidly improved with a change to commercially-pure titanium. After several films were produced and tested, we began elevated temperature testing and discovered that the adhesion of the films was damaged by exposure to elevated temperature. Because the deposition process takes place at elevated temperature, increasing the temperature from room temperature was expected to relieve rather than induce thermal stresses at the interface. Given the unsuccessful experiment with titanium, films deposited on tungsten carbide substrates were produced to be used in the impact-echo sensors.

4.2 Transducer Physical Design

Based on the preliminary tests, the robust impact-echo transducer diagrammed in Figure 3 was designed. A stainless steel case (3) holds the AlN film and substrate (2) which is pushed against a conical tip (1) through a metal foil to create the same architecture as in the feasibility trials. Belleville springs made from inconel (6) push against an alumina insulator (5) and a stainless steel rod (4) to hold these parts tightly together. A tube fitting (7) holds a mineral-insulated armored cable (9) whose center conductor (8) is electrically connected to the substrate (2), forming a signal electrode. The ground electrode is provided by the body of the transducer and the outside of the armored cable. The cable conveys the signal outside the hot area of the test

where it can be connected to an amplifier. Figure 4 shows a photograph of the assembled transducer. The transducer was designed for continuous use up to at least 600°C. Use at temperatures up to 700°C would probably not cause significant damage, but might cause some creep relaxation in the inconel springs. All the other elements, including the piezoelectric film, should both survive and operate at high temperature.

Figure 3. AlN Impact-Echo Transducer Schematic.

Figure 4. AlN Impact-Echo Transducer.

Initial tests with the transducer immediately revealed an operating problem. An internal resonance was causing significant voltage output at a frequency near 10 kHz, regardless of the test object or if the transducer was directly excited by tapping on the tip. Analysis revealed that this was in a range corresponding to a spring-mass resonance of the internal rod and Belleville springs, even though the internal components are compressed tightly against the end of the transducer by the springs and should not have moved when excited. To test this theory, the springs were replaced with a stack of washers, making the internal construction of the sensor essentially solid. This change removed the resonance, but introduced questions as to how a compressive preload would be maintained to couple the film to the conical front end. However, a functioning transducer had been created which was able to acquire signals from concrete test slabs constructed to mimic the refractory slabs to be tested.

Section 5

Impact-Echo and Refractory Material

While transducers were being developed, researchers at the University of Missouri-Rolla had been further testing a commercial impact-echo sensor, as well as acquiring larger slabs of AZS refractory material. The slabs acquired were each 400 mm by 400 mm, with thicknesses of 40, 50, 60, 70, and 80 mm. Room temperature experiments on these slabs with the commercial impact-echo transducer did not result in impact-echo data which could be correlated with thickness. Additional testing performed with the AlN transducers described above gave similar results to the initial testing, where the acquired signals were similar to those from the commercial transducer, although at lower amplitudes. The conclusion was that impact-echo testing was not straightforward for this very dense ceramic material.

One possible reason for the difficulty is that undesirable modes of vibration were excited in the test slab, even though their large aspect ratios were chosen to avoid this problem. A second possibility is that the test object was not adequately excited during testing; this could have occurred if its resonance frequency was above the range of most of the input energy provided by the impactors. A combination of small-diameter, high-mass (tungsten carbide) impactors with high impact velocities was used to provide high-frequency excitation, with no better results than the more commonly-used steel spheres. It is possible that the high sonic velocity of the AZS would allow impact-echo testing if a much larger sample of the material could be used.

Section 6

Alternative Approaches

6.1 Transducer Behavior at Elevated Temperature

Given that there was some hope that the AlN transducers could be used eventually, the next step was to proceed to test the transducer at elevated temperature. For the testing, the transducer body was heated directly and then applied to a test object which was struck with an impactor, as before. Results were again disappointing and the transducers failed to produce usable signals when heated. This failure is partly due to a disruption of the compressive preload on the AlN film by thermal expansion of the transducer housing. Thermal expansion was no longer compensated by the high-temperature springs which were removed, as noted above, to resolve an internal resonance. A redesign with a much heavier internal mass between the springs and the AlN film might lower the resonance below a few kHz and resolve this problem. A second problem was a steady decrease in signal strength, likely caused by charge leakage, accompanied by an increase in the background noise on the output from the sensor. It appeared that the sensor would only be useful in its present form to a maximum of 200°C or less, well below the desired operating range.

6.2 Ultrasonic Evaluation of Refractory

One additional possibility for the use of the AlN sensor for refractory wall thickness measurement existed: traditional ultrasonic evaluation might be feasible because the refractory material is very dense^[4]. This investigation was begun at room temperature on the AZS test slabs using a conventional UTEX 340 ultrasonic instrument with standard, commercially-available transducers. The echo of the ultrasonic pulse from the back wall was easily detectible. While not an ideal material for ultrasonic evaluation, the fuse-cast AZS did transmit sonic energy at frequencies above 1 MHz. Sonic energy above 10 MHz attenuated quickly in the material, most likely due to scattering from ceramic grains and porosity. In order to eliminate front surface echoes and maximize the useful signal, a pitch-catch arrangement of two transducers was used for the experimentation. This arrangement also facilitated the introduction of buffer rods between the transducers and the refractory, critical for elevated temperature measurements with traditional commercial transducers.

The AlN transducers did not require a buffer rod for thermal protection, but did require a front face to maintain the integrity of the transducer. Two of the impact-echo transducers were modified to an ultrasonic configuration. One modification simply consisted of removing the conical tip and grinding the face of the transducer flat to provide a coupling surface. A more complex modification was attempted for the second transducer, drilling through the front surface and press-fitting an alumina rod to provide coupling to the test object and integrity to the transducer. The additional transducer, shown in Figure 5, was also constructed to have a thinner steel front face.

Figure 5. Thin-faced "pipe" transducer.

For elevated temperature testing, a 40 mm thick AZS slab replaced the lid of a furnace. Thermocouples were used to monitor both the hot and cold faces of the slab. Ultrasonic data were collected at four points on the slab, whose horizontal top greatly simplified the manipulation of transducers. Preliminary experiments at room temperature showed that measurements could be taken using a Panametrics 3.5 MHz conventional transducer to send ultrasonic energy, and either a conventional or AlN transducer to receive it. These experiments were performed using a water-based couplant gel. Figure 6 shows the test configuration using a conventional transducer with a buffer rod and an AlN transducer. As the temperature increased, other couplants were required at the hot side of the buffer rod^[4]. Commercial high-temperature couplants from Panametrics and Sonotech performed well up to 275°C and marginally up to 400°C. Various metal foils and graphite foil were tried above that temperature with no success. At high temperature, alumina, mica-glass ceramic, and steel buffer rods were placed between the conventional transducers and the test piece, with permissible contact times of less than a minute before removing them for cooling.

Figure 6. Square AlN transducer on AZS slab next to buffered conventional transducer.

The AlN transducer did not require a buffer rod, but did require couplant. The AlN transducer with the ceramic rod face was too difficult to balance manually while maintaining tight coupling, so efforts focused on the metal-faced AlN transducers. These transducers had center frequencies in the 20-30 MHz range, and therefore were used only as receivers because ultrasonic pulses produced by them did not contain enough lower-frequency (<5 MHz) energy to penetrate the refractory test object effectively. Figure 7 shows an ultrasonic pulse sent by a conventional transducer into a 50 mm AZS slab, reflected off the back wall and received by an AlN transducer at room temperature. Note that, while the energy produced by the conventional transducer was centered at 3.5 MHz, the received pulse width indicates energy centered at 11 MHz. The majority of the interrogating pulse energy is, thus, not sensed efficiently by the high-frequency AlN transducer. When coupled with the decrease in signal-to-noise ratio as the temperature was increased, this lower sensitivity made the AlN transducers useful only to 275°C. Figure 8 shows ultrasonic waveforms from the AlN and conventional transducers near 120°C. Note that the addition of the buffer rod and temperature effects has forced the use of higher gain than at room temperature, and that the AlN signal is 10dB lower amplitude. Figure 9 shows ultrasonic waveforms from the AlN and conventional transducers near 220°C, where the AlN pulse is misshapen. At 275°C in Figure 10, the pulse shape has returned to more nearly normal, but the amplitude has become much smaller. Upon further temperature increases, the signal from the AlN transducer became lost in the noise. The conventional transducer produced useful data to about 500°C, where the couplants became useless and allowable contact times became very small.

Figure 7. Pulse through 50 mm AZS slab at room temperature.

Figure 8. Pulses through 40 mm AZS slab near 120°C surface temperature.

Figure 9. Pulses through 40 mm AZS slab near 220°C surface temperature.

Figure 10. Pulse through 40 mm AZS slab at 275°C surface temperature.

Section 7

Conclusions and Recommendations

The most important conclusion to be drawn from this project is that the use of high-temperature AlN transducers and an impact-echo method to monitor refractory wall recession does not appear to be practical. First, the impact-echo method itself seems to have difficulty with refractory materials such as AZS, and is likely to become imprecise during the end of life of a refractory, when metal line erosion approaches the outer wall, temperature gradients increase, and the structure no longer resembles a plate. Second, the AlN transducers were inefficient for the reception of impact-echo energy, being more than 10 dB less sensitive than a buffered commercial transducer at slightly-elevated temperature and displaying declining sensitivity with increased temperature.

The ultrasonic method shows more promise, but several barriers stand in the way of practical use. If conventional transducers are to be used, buffer rod and couplant issues add tremendous complexity to the application. The use of laser-based ultrasound^[5], while expensive, might be justified for critical areas. Aluminum nitride transducers in direct contact with the hot test article will likely be a future alternative. The transducers in this program could not work at the low frequencies required; in fact, piezoelectric AlN films of the thickness required for such frequencies cannot be practically produced at this time. However, the electronics industry is developing the ability to grow large single crystal AlN. Thick (~1 mm) pieces of such a crystal could operate at the required frequencies. Ultrasonic coupling remains a problem at elevated temperatures, but the use of metal foils or molten glasses has been documented. Thus, it appears that direct ultrasound, enabled with high-temperature piezoelectric transducer such as AlN, is a more promising technology than impact-echo for this application, but much development remains.

Section 8

References

1. Sansalone, Mary and Streett, William. *Impact-Echo: Non-destructive Evaluation of Concrete and Masonry*. Ithica, NY: Bullbrier Press, 1997.
2. Proctor, T.M., Jr., “More recent improvements on the NBS conical transducer”, J. Acoustic Emission 5 (4), 1986, pp. 134-142.
3. *Acoustic Emission Testing: Nondestructive Testing Handbook* (2nd ed.) v.5. Paul McIntire, ed., Columbus, OH: American Society for Nondestructive Testing, 1987, pp. 575-585.
4. Fleischmann, Bernhard. “Ultrasonic Determination of the Residual Wall Thickness of Glass Furnace Refractories”, Glastech. Ber. Glass Sci. Technol. 70 (1997) No.1.
5. Matsumoto, Takahiro, et.al., “Measurement of High-Temperature Elastic Properties of Ceramic Using a Laser Ultrasonic Method”, J. Am. Ceram. Soc., **84** [7], 2001, pp.1521-25.