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Abstract. As part of the Advanced Test Reactor National Scientific User Facility (ATR-NSUF) program, the Idaho National Laboratory (INL) has developed in-house capabilities to fabricate, test, and qualify new and enhanced temperature sensors for irradiation testing. Clearly, temperature sensor selection for irradiation tests will be determined based on the irradiation environment and budget. However, temperature sensors now offered by INL include a wide array of melt wires in small capsules, silicon carbide monitors, commercially available thermocouples, and specialized high temperature irradiation resistant thermocouples containing doped molybdenum and niobium alloy thermoelements. In addition, efforts have been initiated to develop and evaluate ultrasonic thermometers for irradiation testing. This array of temperature monitoring options now available to ATR and other Material and Test Reactor (MTR) users fulfills recent customer requests.

Keywords: Advanced Test Reactor, Irradiation Testing, Temperature Monitoring.
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INTRODUCTION

An increasing number of U.S. nuclear research programs are requesting enhanced in-pile instrumentation that can provide real-time measurements of key parameters during irradiations. Additionally, fuel cycle research and development (FCR&D) efforts funded by the U.S. Department of Energy (DOE) now emphasize approaches that rely on first principle models in order to develop improved fuel designs that offer significant improvements over current fuels. To facilitate this approach, high fidelity, real-time data of unprecedented accuracy and resolution are essential for characterizing the performance of new fuels during irradiation testing. Furthermore, sensors that obtain such data must be miniature, reliable and able to withstand high temperature, irradiation conditions. Depending on user needs, sensors may need to obtain data in inert gas, pressurized water, or liquid metal environments.

One of the facilities in which DOE fuel irradiations are performed is the Advanced Test Reactor (ATR). The DOE designated the ATR at the Idaho National Laboratory (INL) as a National Scientific User Facility (NSUF) in April 2007 to support U.S. research in nuclear science and technology. A key component of the ATR NSUF effort is to develop and implement in-pile instrumentation capable of providing real-time measurements of key parameters during irradiation.

As a result of this ATR NSUF effort, INL now offers several new temperature sensors to users at the ATR and other Material and Test Reactors (MTRs). In addition, INL has initiated efforts to develop an ultrasonic thermometry capability for irradiation testing. This paper describes the various temperature options available and under development. In addition, a brief description of the ATR and typical irradiation locations is provided to illustrate typical conditions where these sensors are deployed. Also discussed are current efforts to develop an ultrasonic thermometry capability for use at ATR and other MTRs.

ATR Design and Irradiation Capabilities

The ATR is a versatile tool for conducting nuclear reactor, nuclear physics, reactor fuel, and structural material irradiation experiments [1].

The ATR's maximum power rating is 250 MW_{th} with a maximum unperturbed thermal neutron flux of 1 $x10^{15}$ n/cm^2-s and a maximum fast flux of 5 x 10^{14} n/cm^2-s.

As shown in Figure 1, the ATR core consists of 40 curved plate fuel elements in a serpentine arrangement around a 3 x 3 array of primary testing locations, including nine large high-intensity neutron flux traps. In addition to the nine large volume (up to 1.22 m long and up to 0.13 m diameter) high-intensity neutron flux traps. There are 66 irradiation positions inside the reactor core reflector tank, and two capsule irradiation tanks outside the core with 34 low-flux irradiation positions.

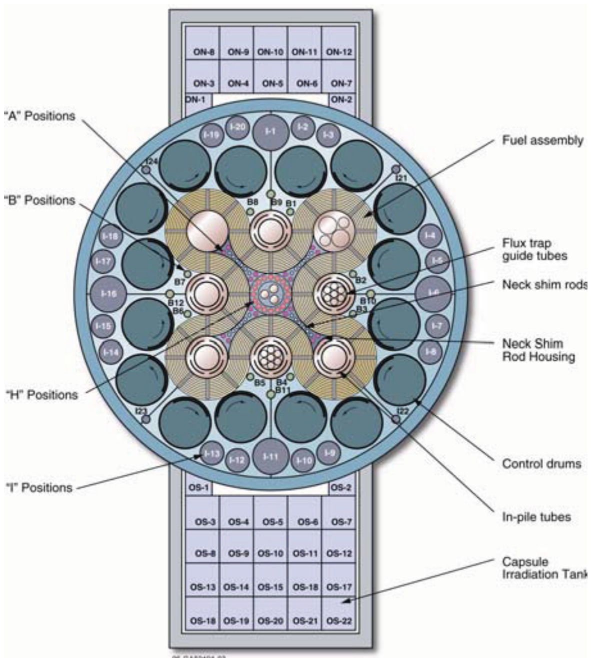

Figure 1. ATR core cross section showing irradiation locations.

There are several ATR test assembly configurations:

• Static capsules may contain several small samples or engineered components. Static capsule experiments may be sealed or may open to ATR primary coolant. Irradiation temperature may be selected (within limits) by providing a gas gap in the capsule with a known thermal conductance. Prior to the current effort, temperature monitoring for such locations was limited to sensors that detect peak temperature such as melt wires.

• Instrumented lead experiments provide active control of experiments and data from test capsules during irradiation using core positions with instrumentation cables. In addition to the sensors used in static capsules, these experiments can use thermocouples for real time temperature monitoring.

• Five of the nine ATR flux traps used for materials and fuels testing are equipped with pressurized water loops (at the NW, N, SE, SW, and W locations) capable of replicating the conditions of a pressurized water reactor (PWR). These loops can operate above the standard temperatures and pressure of a commercial PWR power plant. Sensor options are the same as for an instrumented lead test.

• The Hydraulic Shuttle Irradiation System (HSIS) or "rabbit" enables insertion and removal of experiment specimens during ATR during operational cycles. Sensor options are the same as for a static capsule test.

IMPROVED ATR TEMPERATURE SENSOR OPTIONS

As discussed in this section, ATR NSUF efforts have resulted in several enhanced temperature monitoring options are now available at the ATR and offered to users of other MTRs.

Encapsulated Melt Wires

Melt wires are the simplest and least expensive option available for monitoring temperature in ATR tests. Metallic wires of a known composition and well characterized melting temperature are included in a test to determine if a specific peak temperature has been reached or exceeded. As noted in ASTM standard E 1214-06 (Reference 3), melt wire materials should consist of metals with 99.9% purity or be eutectic alloys such that their melting temperatures are within ± 3 °C of recognized values. The standard also states that transmutation-induced changes of these wires are not considered significant up to 1×10^{20} n/cm^2 (E > 1 MeV). As noted in Reference 3, melt wires should be selected to measure temperature at 5 to 12 °C intervals, with at least one melt wire that possesses a melting temperature greater than the highest anticipated temperature.

INL has in-house capabilities to verify the melting temperature of candidate wire materials (ranging from ~85 to 1500 °C) using a differential scanning calorimeter (DSC) installed at INL's High Temperature Test Laboratory (HTTL). INL also has a new capability to encase multiple melt wires into a single 1.6 mm diameter quartz capsule, such as the one shown in Figure 2, for irradiation testing. Figure 2 shows a single melt wire capsule after progressive heating steps during verification testing in high temperature furnaces installed at INL's HTTL. The wires selected are chosen based on expected irradiation test temperatures and required resolution. When possible, INL selects high purity materials with low thermal neutron capture cross-sections.

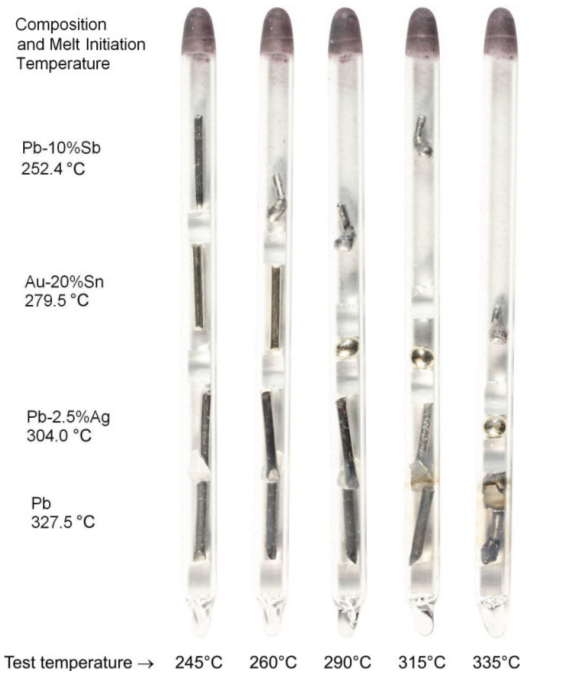

FIGURE 2. Quartz tube containing four melt wires in separated compartments.

The melt wires depicted in Figure 2 are considered good materials for this purpose because melting may be determined by visual inspection; that is, it is possible to judge if the wires have melted by inspecting the capsule with the naked eye. This is important because this determination must be made during Post Irradiation Examination (PIE) in a hot cell. Figure 3 shows melt wires that were determined to be non-ideal choices for use because melting (confirmed through DSC testing) could not easily be determined by visual inspection; and microscopic examination was required. Melting in these cases is determined by changes to the surface condition of the wires rather than bulk deformation of the wires.

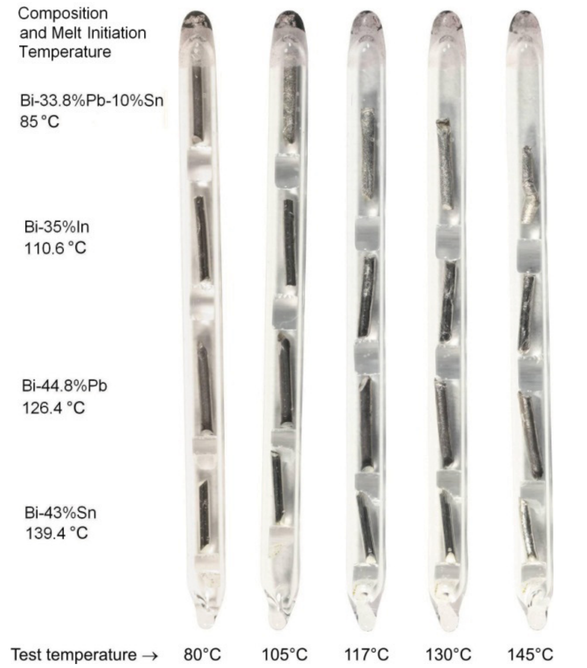

FIGURE 3. Quartz tube containing non-ideal melt wires.

Table 1 lists the melt wire materials currently available for irradiation testing that have undergone verification testing.

Table 1. Melt Wire Materials and Melting Temperatures in INL Library

Material (wt% of components)	Melt Onset, °C
56.2Bi 33.8Pb 10Sn	85.0
65Bi35In	110.6
55.2Bi 44.8Pb	126.4
57Bi43Sn	139.4
100Sn	231.8
95Sn/5Sb	238.6
90Pb/10Sb	252.4
80Au 20Sn	279.5
90Pb 7.5Sn 2.5Ag	290.0
97.5Pb5Ag5Sn	302.9
97.5Pb2.5Ag	304
97.5Pb1 .75Sn 1.75Ag	309.3
100Pb	327.5
100Zn	419.6
100Al	660.5
49Ag16Cu23Zn7.5Mn4.5Ni	681.3
40Ti20Zr20Cu20Ni	850.7
98.2Cu1.8Be	865.1
100Ge	938.3

Material (wt% of components)	Melt Onset, °C
82Au 18Ni	955.0
100Ag	961.9
65Cu35Au	995.6
100Au	1064.0
100Cu	1084.6
70Cu30Ni	1191.0
28Mo69Ni2Fe1Co1Cr	1370.0
100Ni	1455.0

Metallic tubes are also available for use in ATR tests. However, post-test examination is more complicated, requiring radiography, computed tomography, or destructive examination of the tube to allow inspection of the wire.

Silicon Carbide Temperature Monitors

Another option available to ATR users is a silicon carbide (SiC) temperature monitor. The benefit of this option is that a single small monitor (typically 1 mm x 1 mm x 10 mm or 1 mm diameter x 10 mm length) can be used to detect peak irradiation temperatures between 200 and 800 °C. SiC incurs damage to its crystalline structure under irradiation. This causes several measureable physical changes to the monitor (i.e. changes in dimensions, thermal conductivity, electrical resistivity, etc.). When the monitor is annealed at a temperature greater than the peak irradiation temperature, the altered properties rapidly change. [4,5,6]

Snead et al. at Oak Ridge National Laboratory (ORNL) [7] successfully used changes in electrical resistivity to detect peak irradiation temperatures with accuracies of approximately 20 °C for dose ranges of 1 to 8 dpa and temperatures between 200 and 800 °C.

Specialized equipment at INL's HTTL now allows peak temperature detection using previously-irradiated SiC monitors. In this technique, the SiC sensor electrical resistivity is measured after annealing in a furnace located within a stainless steel enclosure at the HTTL (shown in Figure 3). After annealing, cooled samples are placed into a constant temperature environmental test chamber to ensure electrical resistivity measurements are taken within 0.2 °C of a predetermined temperature, 30 °C. A high accuracy (9 digit) multimeter, located outside the stainless steel enclosure, is used to obtain resistance measurements. Specialized fixturing (Figure 3) was developed to ensure that measurements are taken with the SiC sensors placed consistently in the same orientation. A four point probe technique is used with the points connected to the sample through spring-loaded, angled electrodes that hold the SiC temperature monitor in place. Current and voltage are provided to the sample via wires that are threaded through the holes in the electrodes.

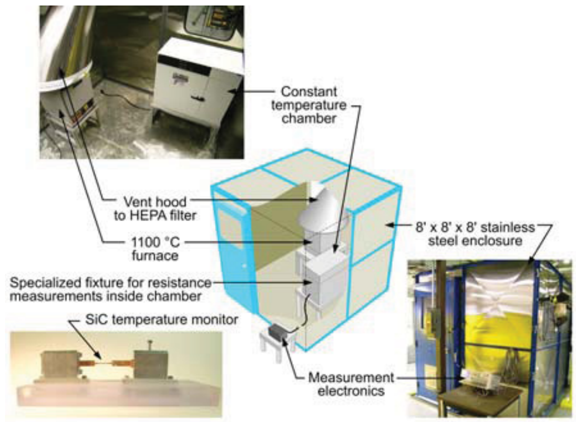

FIGURE 3. Setup to anneal and measure electrical resistivity of SiC temperature monitors.

The accuracy of this new INL capability was verified by completing comparison measurements with ORNL on identical sensors that had been subjected to identical irradiation conditions [8]. Each sensor was broken into two pieces, with one piece tested at ORNL and the other at INL. Results, as shown in Figure 4, indicate that similar peak irradiation temperatures are inferred from ORNL and INL measurements for such cases. Results from both laboratories indicate that there is a significant increase in the normalized electrical resistivity at 670 °C.

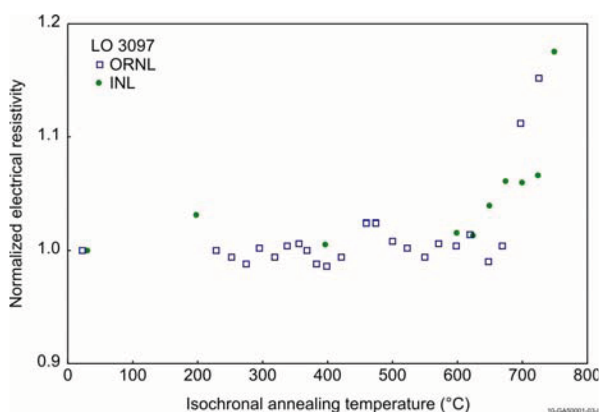

FIGURE 4. Electrical resistivity measurement comparison on SiC monitors irradiated at peak temperature of 670 °C.

High Temperature-Irradiation Resistant Thermocouples

Real time temperature monitoring is typically accomplished using thermocouples. Type K or Type N thermocouples have demonstrated excellent reliability and signal stability under irradiation, even for very high neutron fluences exceeding 10^{22} n/cm^2 (thermal neutrons). However, these thermocouples decalibrate when used at temperatures above 1100 °C. Although other types of commercially-available thermocouples exist for higher temperature applications (up to 1800 °C), the thermoelements used in these thermocouples contain materials such as tungsten and rhenium (in the case of Type C and D thermocouples) or platinum and rhodium (Type R, S, and B) that rapidly decalibrate due to transmutation from absorption of neutrons. A High Temperature Irradiation Resistant ThermoCouple (HTIR-TC) was developed at the HTTL that contains commercially-available doped molybdenum paired with niobium alloy thermoelements.

Materials for HTIR-TC components were selected based on results of high temperature materials interaction tests, ductility investigations, and temperature response evaluations ([9] - [13]).

HTIR-TC long duration performance has been demonstrated through testing, in which thermocouples were heated to temperatures ranging from 1200 °C to 1800 °C and held at temperature for up to 6 months. The 1200 °C test included nineteen commercially-available Type N thermocouples, three commercially-available Type K thermocouples, and nine INL-developed HTIR-TCs. As indicated in Figure 5, some of the tested Type K and N thermocouples were observed to drift by over 100 °C or 8% while significantly smaller drift (typically less than 20 °C or 2%) was observed in the HTIR-TCs. Similar drifts (2%) were observed in HTIR-TCs during a long duration (4000 hour) test completed at 1400 °C, and smaller drifts (less than 1%) have been observed in HTIR-TCs with enhanced fabrication techniques for higher temperatures (up to 1800 °C).

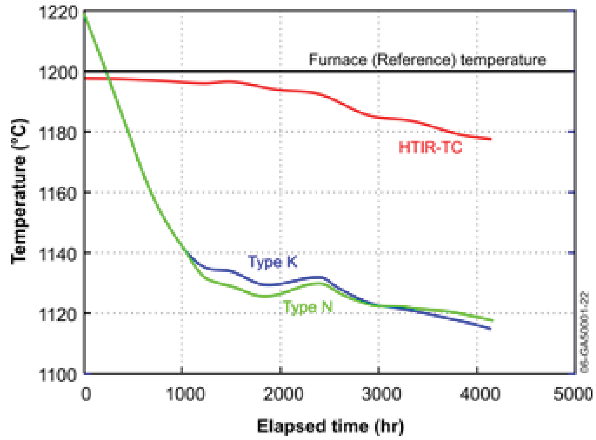

FIGURE 5. Representative signal drift of test thermocouples in long duration INL 1200 °C tests.

HTIR-TCs have also been tested for long periods while exposed to neutron radiation. These TCs were installed in a multi-capsule gas reactor fuel experiment in which samples were irradiated at temperatures up to 1200 °C in the ATR. The test ran for 2 years and 8 months. Figure 6 shows signals from two HTIR-TCs and one commercial Type N thermocouple located within one of the capsules. Signal variations are due to ATR power fluctuations and outages (e.g., gray regions correspond to periodic ATR shutdowns). As shown in Figure 6, the HTIR-TC (designated TC-4-1) located near the Type N thermocouple (designated TC-4-3) yielded a signal consistent with the signal from this Type N thermocouple at the beginning of this irradiation. In addition, the HTIR-TC located at a higher temperature region within the capsule (designated TC-4-2) yielded a consistent, but higher temperature, signal. In October 2008, the Type N thermocouple failed; and its signal ceased. However, the two HTIR-TCs continued functioning throughout the duration of the test.

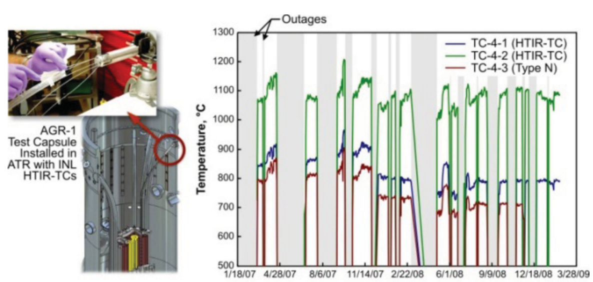

FIGURE 6. HTIR-TCs installed in AGR-1 test capsule and representative HTIR-TC and Type N data during ATR irradiation.

The successful operation of HTIR-TCs in this test has led to requests for INL to supply them to other MTRs. In 2010, HTIR-TCs were supplied to the Massachusetts Institute of Technology (MIT) for an irradiation test in the MIT Nuclear Research Reactor (MITR). In addition, HTIR-TCs have recently been fabricated for the Halden Boiling Water Reactor (HBWR). These HTIR-TCs were shipped to the Institute for Energy Technology at the Halden Reactor Project (IFE/HRP) in November 2011.

Ultrasonic Thermometry

INL is currently in the process of developing an ultrasonic thermometry capability. Ultrasonic Thermometers (UT) 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 for irradiation testing purposes is the pulse echo method. By sending an ultrasonic pulse through a thin rod, and measuring the time between the initial pulse and the reflection of the pulse from the opposite end of the rod, the average acoustic velocity (and, therefore, the average temperature of the rod) may be calculated. By introducing acoustic discontinuities, such as notches or sudden diameter changes, the rod may be segmented into a multipoint temperature sensor in which 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 acoustic wavelength, which historically has led to UT diameters less than 1.6 mm [14]. If this maximum diameter condition is met, the temperature-dependant acoustic velocity, $v(T)$, is related to density, $\rho(T)$, and elastic modulus, $E(T)$, (both of these properties are also temperature dependant) through the following equation:

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

In addition to their small diameters, UT's offer several potential advantages over other temperature sensors. Measurements may be made near the melting point of the sensor material, allowing monitoring of temperatures potentially in excess of 3000 °C. In addition, because no electrical insulation is required, shunting effects (often problematic for thermocouples at temperatures above 1800 °C) are avoided. Most attractive, however, is the ability to introduce multiple acoustic discontinuities to the sensor, as this enables temperature measurements at several points along the sensor length.

A conceptual design of a typical multi-sensor UT system, with key components identified, is shown in Figure 7. 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 portion of the pulse energy is reflected at each discontinuity (notches, diameter changes, etc.). Each reflected pulse is received by the excitation coil, transformed into an electrical signal, then 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.

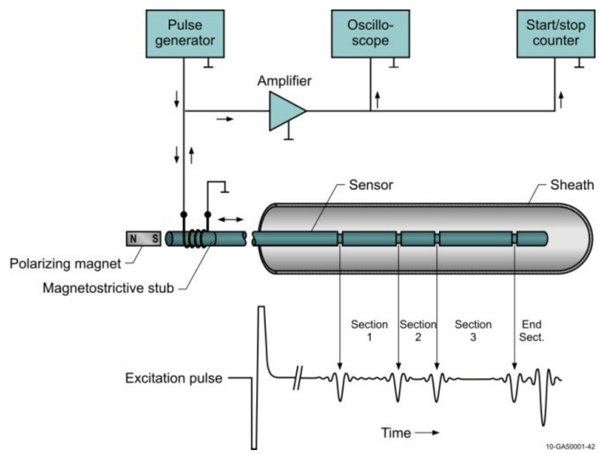

FIGURE 7. A typical multi-segment pulse/echo ultrasonic thermometry system.

The current research is focused on improving three key aspects of UT performance that have hindered the application of UTs to irradiation testing. The first task is selecting appropriate probe materials. Prior work focused on measurements of extremely high temperatures, such as those reached during a fuel melt event [15,16]. Metals with very high melting temperatures, such as tungsten (3410 °C) and rhenium (3186 °C), were used. Both of these metals have relatively high thermal neutron capture cross-sections (18.3 barns for tungsten and 89.7 for rhenium), and tend to decalibrate significantly over the course of an irradiation test. Other refractory metals with lower neutron capture cross-sections, such as molybdenum (2623 °C melting point and 2.6 barn cross section), or newer materials, such as high temperature ceramics, may offer superior performance to previously tested materials, while reducing the detrimental effects of the reactor environment.

The signals generated and detected by an ultrasonic thermometry system are complex and noisy, due to the multiple echoes produced by the acoustic discontinuities that define the sensor segments and the signal attenuation that can occur at high temperatures. As such, improvements must be made to the signal processing system. Specifically, improvements must be made to enhance the signal to noise ratio and the ability to discriminate between primary and subsequent echoes.

The third improvement required for the successful deployment of UT's in irradiation testing is the elimination of sticking. Sticking refers to contact bonding of the sensor with its solid surroundings (e.g., a sheath, fuel, cladding, etc.). At high temperatures, diffusion processes cause the sensor to become attached to the surrounding material. This causes an acoustic impedance mismatch that generates spurious echoes which can obscure the echoes used for temperature measurement. If the sticking is significant, the signal may be completely lost. Previous attempts at reducing sticking included reducing contact between the sensor and sheath by introducing standoffs to the sensor or by reducing the diffusion potential through the use of coatings or different sheath materials. Standoffs include metallic spheres welded to the sensor, coils of wire wrapped around the sensor, and (most successfully) a fine tungsten cloth wrapped around the sensor. Typically, reducing the diffusion potential involved using a thoria sheath or coating. All of these methods had limited success, but no method to date has eliminated sticking completely.

SUMMARY

Several improved methods of temperature monitoring have recently been made available to ATR users through development efforts at INL's HTTL. The least expensive is the multiple melt wire capsule, which gives an estimate (based on the melting temperatures of the wires) of the maximum temperature reached during a test. Silicon carbide monitors are not limited to indicating whether a single temperature has been exceeded, but can cover a range of temperatures with a single monitor. For real time monitoring, INL-developed HTIR-TCs can be used. These thermocouples offer a higher temperature capability than commercially available thermocouples such as Type-K and Type-N thermocouples. HTIR-TCs are not sensitive to neutron induced decalibration, unlike tungsten/rhenium thermocouples. INL is also developing an ultrasonic thermometry capability that will offer ability high temperature option to measure a temperature profile, in real time, with a single small diameter sensor.

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