Hot Wire Needle Probe for In-Reactor Thermal Conductivity Measurement

Joshua E. Daw, Joy L. Rempe, and Darrell L. Knudson

***Abstract*— Thermal conductivity is a key property that must be known for proper design, test, and application of new fuels and structural materials in nuclear reactors. Thermal conductivity is highly dependent on the physical structure, chemical composition, and the state of the material. Typically, thermal conductivity changes that occur during irradiation are measured out-of-pile using a "cook and look" approach. Repeatedly removing samples from a test reactor to measurements is expensive, has the potential to disturb phenomena of interest, and only provides understanding of the sample's end state when each measurement is made. There are also limited thermophysical property data for advanced fuels. Such data are needed for simulation design codes, the development of next generation reactors, and advanced fuels for existing nuclear plants. Being able to quickly characterize fuel thermal conductivity during irradiation can improve the fidelity of data, reduce costs of post-irradiation examinations, increase understanding of how fuels behave under irradiation, and confirm or improve existing thermal conductivity measurement techniques. This paper discusses efforts to develop and evaluate an in-pile thermal conductivity sensor based on a hot wire needle probe. Testing has been performed on samples with thermal conductivities ranging from 0.2 to 22 W/m·K at temperatures ranging from 20 °C to 600 °C. Thermal conductivity values measured using the needle probe match data found in the literature to within 5% for samples tested at room temperature, 6% for low thermal conductivity samples tested at high temperatures, and 10% for high thermal conductivity samples tested at high temperatures.**

***Index Terms*— In-pile instrumentation, nuclear fuel properties, thermal conductivity measurement.**

I. Introduction

THERMAL properties of materials must be known for proper design, test, and application of new fuels and structural materials in nuclear reactors. In the case of nuclear fuels during irradiation, the physical structure and chemical composition change as a function of time and position within the rod. There are limited thermal property data for advanced fuels. Such data are needed for simulation design codes, used in the development of next generation reactors. Being able to quickly characterize fuel thermal conductivity during irradiation can reduce costs from PIE examinations, increase understanding of how fuels behave under irradiation, and confirm or improve existing thermal conductivity measurement techniques.

An effort has been initiated by the Idaho National Laboratory (INL) to investigate the viability of an in-pile thermal conductivity probe based on the Transient Hot Wire Method (THWM), which is an adaptation of the American Society for Testing and Materials (ASTM) needle probe method [1]. The needle probe method is based on the theory of an infinite line heat source applying a constant heat flux to a semi-infinite solid. The probe contains a linear heating element and a temperature sensor inserted into a material whose thermal conductivity is to be measured. The thermal conductivity is calculated from the recorded thermal response of the sample. Preliminary investigations by INL indicate that this approach may offer advantages over steady state techniques [2].

A. Background

Measuring thermal conductivity of materials during irradiation poses significant challenges. Any sensor used in-situ must be robust, as it is subjected to very high temperatures, high pressures, and thermal and fast neutron fluxes. Compatibility between the probe and the test environment is ensured primarily through selection of appropriate materials. These materials must have high melting temperatures, low neutron absorption cross sections, and they must not interact at high temperatures. The sensor also must be minimally intrusive, such that it has a minimal effect on the measurement.

Historically, fuel thermal conductivity has been measured using either Post Irradiation Examination (PIE) measurements or in-situ using a two thermocouple method based on Fourier's Law of heat conduction for a solid with internal heat generation [3]. Repeatedly removing samples from a test reactor to make out-of-pile measurements during PIE is expensive, has the potential to disturb phenomena of interest, and only provides understanding of the sample's end state at the time each measurement is made. The two thermocouple method uses one or more thermocouples inserted near the center of the fuel rod and one exterior to the fuel (in the coolant or a structure outside the fuel element). Thermal conductivity is derived from the temperature difference between the two thermocouples at steady-state. Currently, the Halden Boiling Water Reactor (HBWR) is the only test reactor where in-pile fuel thermal conductivity measurements are still performed [4]-[6]. The Institute for Energy Technology at Norway's Halden Reactor Project (IFE/HRP) has used this technique to assess the effect

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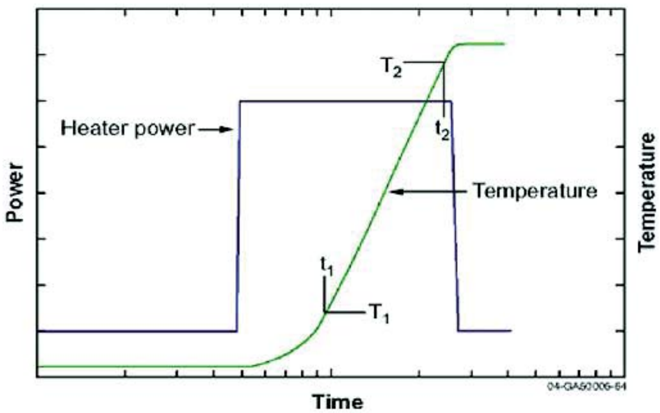

Fig. 1. Semi-log temperature rise plot for transient methods.

of burnup on thermal conductivity. However, this method requires several assumptions, such as uniform fuel composition, uniform fuel density, minimal gap conductance effects, and uniform heat generation in the fuel rod. IFE/HRP tests are typically performed with specially-designed fuel rods with a small as-fabricated fuel-to-clad gap to minimize the influence of gap conductance change (densification/swelling, fission gas release) on the fuel centerline temperature during irradiation.

B. Principles of Technique

The temperature rise from an internal heat source in a material is dependent on the material's thermal conductivity [7], [8]. In a solid, this method may be applied by embedding a line heat source in the material whose thermal conductivity is to be measured. From a condition of thermal equilibrium (required to ensure the measurement is not affected by temperature changes caused by nuclear heating), the heat source is energized and heats the sample with constant power. The temperature response of the sample is a function of its thermal properties, and the thermal conductivity is calculated from the temperature rise detected in the sample. Following a brief transient period, a plot of the temperature versus the natural logarithm of time becomes linear, as shown in Figure 1 (linear region of the time interval between times t_1 and t_2 and temperatures T_1 and T_2). The slope of the linear region is used to calculate the test material thermal conductivity.

The needle probe method is based on the theory of an infinite line heat source embedded within a semi-infinite solid. In this configuration, the thermal response is detected by a sensor (such as a thermocouple) located a finite distance from the heat source. The needle probe is designed such that the heat source and thermocouple are both located within the probe. A schematic diagram showing components of a thermal conductivity needle probe is shown in Figure 2.

The thermal conductivity of the sample material is derived from the slope of the thermal transient using the following relation from the ASTM needle probe testing standard [1]:

$$k = \frac{CQ}{4\pi LS} \tag{1}$$

where k is the thermal conductivity, C is a calibration factor, Q is power dissipated by the heater, L is the heater length, and

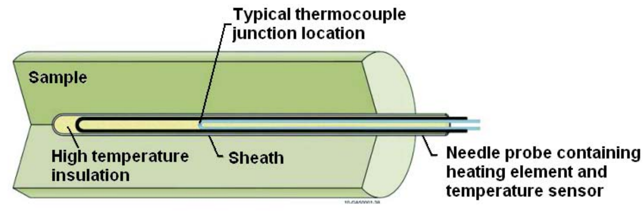

Fig. 2. Schematic diagram of thermal conductivity needle probe.

S is the slope of the linear portion of the transient response. Note that for a small probe (less than 2.5 mm in diameter) C may be neglected. As discussed previously (see Figure 1), the slope of the linear section of the transient response is given by

$$S = \frac{T_2 - T_2}{\ln \frac{t_2}{t_1}}. \tag{2}$$

The length of the heater is a constant, while the temperature and time are recorded. The dissipated power is calculated from the current and voltage applied to the heater.

C. Selection of Time Interval

The theoretical model described in Figure 1 implies that this technique is relatively simple. However, testing reveals that this is not always the case. For materials with low thermal conductivity, the linear segment of the thermal response is easy to identify, as the duration and temperature rise are relatively large. For high conductivity materials, this is not the case; both duration and temperature rise are greatly reduced. This problem is compounded if the sample diameter is reduced. The ability to select a proper time interval is critical for accurate calculation of thermal conductivity as Equation 1 requires that the effect of boundary heat transfer conditions not be reflected in the measured temperature data (including this data will yield inaccurate results). For this reason, a hot wire probe and test sample must be tested under a variety of conditions prior to deployment such that the characteristics of the individual probe may be quantified. For example, the earliest transient portion of the response curve is primarily dependant on the probe characteristics. Each probe must be tested to quantify its time constant (the time required for the transient response of the probe to end), so that the portion of the thermal response due to the probe may be neglected. Identifying the time constant of the probe is accomplished by testing the probe in two or more samples with different thermal conductivities. Figures 3 and 4 show the results of this test performed on two probes (one with a length of 152.4 mm and one with a length of 63.5 mm). The time prior to divergence of the data for different materials is attributable to the response of the probe, approximately 6 seconds for the longer probe and slightly over 1 second for the short probe.

Additionally, thermal contact between the probe and sample has an effect on the measured response. Assuming the sample is of sufficient size, the effect of poor thermal contact can (to an extent) be ignored. Heat buildup in the gap between the probe surface and sample will cause a temperature rise

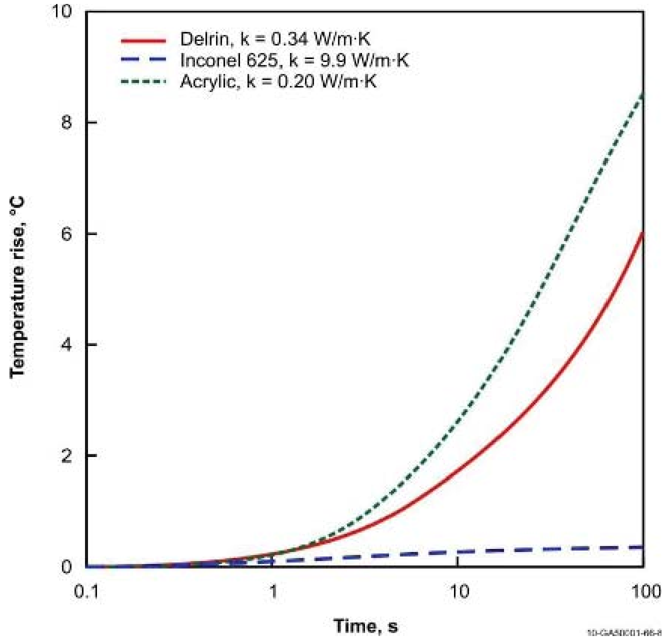

Fig. 3. Test results identifying time constant of a 63.5-mm-long needle probe at room temperature.

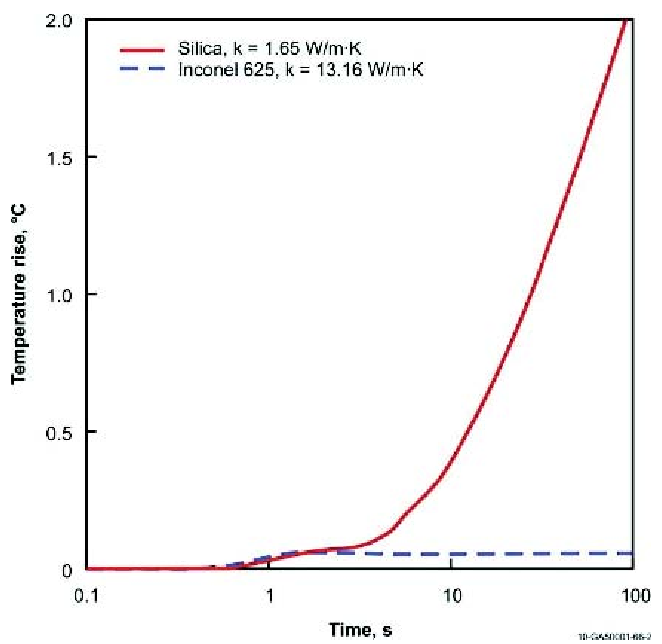

Fig. 4. Test results identifying time constant of a 152.4-mm needle probe at 250 °C.

and delay the onset of linearity, as shown in Figure 5. Note that the data shown represent a probe and sample with thermally conductive grease creating good thermal contact, and a probe and sample without thermal grease but in good contact.

The end of the linear region can also be difficult to identify, as it is strongly influenced by thermal diffusivity, sample diameter (assuming a cylindrical sample), and heat transfer conditions at the outer surface of the sample. Identifying the approximate time of thermal dissipation through the sample yields an upper bound on useful data as well as necessary test

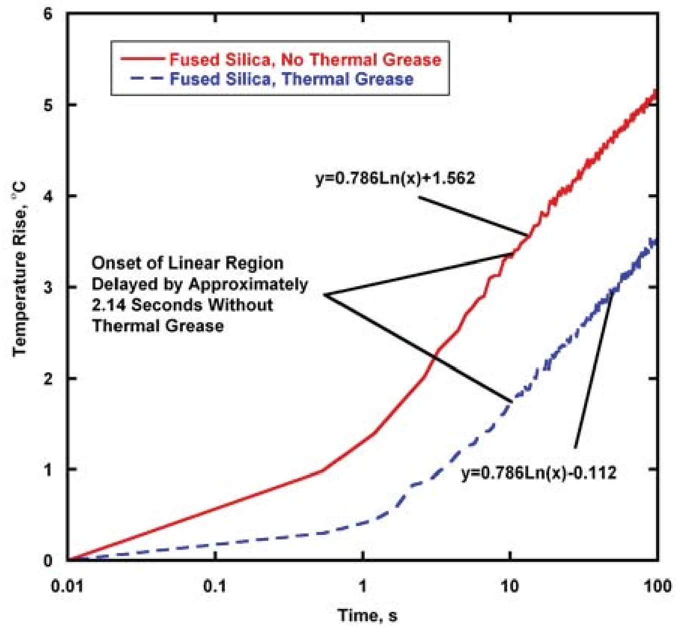

Fig. 5. Effect of “poor” thermal contact between probe and sample.

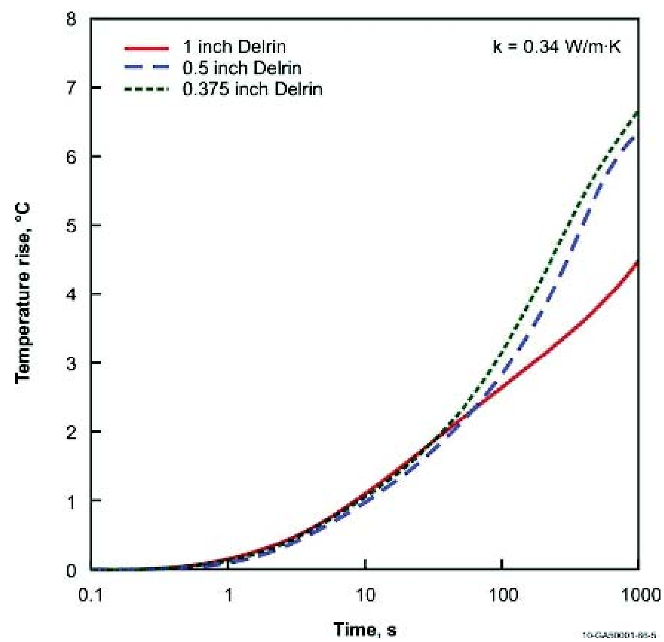

Fig. 6. Test results demonstrating effect of sample diameter.

duration. Figures 6 and 7 demonstrate the effects of varied sample diameters and surface conditions.

In summary, evaluations to evaluate the range of applicability for the hot wire needle probe have led to the following insights:

1) The probe and thermal contact between the probe and sample dominate initial temperature response. Thermal conductivity evaluations must only consider data obtained after the probe response time.
2) Shorter test durations must be used in tests with the following conditions:
   a) smaller diameter samples
   b) higher thermal conductivity samples

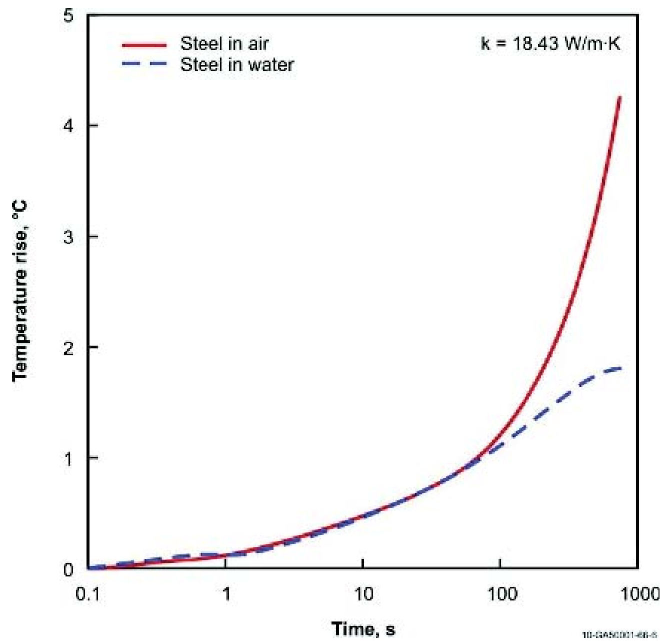

Fig. 7. Test results demonstrating effect of surface conditions.

TABLE I
ROOM TEMPERATURE TESTING RESULTS

Material	INL average (W/m•K)	Reported (W/m•K)	% Difference
Fused Silica	1.4	1.4 [10]	0
304L Stainless steel	15.9	15.3 [10]	3.5
Titanium-6%Al-4%V	7.3	7.2 [11]	1.3
Inconel 625	10.3	9.9 [12]	3.7
Delrin	0.33	0.34 [13,14]	2.9
Acrylic	0.21	0.20 [15]	5.0

3) Thermal conductivity evaluations must be completed prior to when the boundary conditions begin to affect the temperature response

Due to these results, it is evident that there is a minimum required sample size that is dependent on the thermal diffusivity of the sample material. That is, if the sample is too small, the response will not fully develop before surface conditions begin to affect the response. This may happen even before the probe response has subsided.

II. PRELIMINARY RESULTS

A. Review Stage

The following section details results of preliminary tests that have been carried out in order to test the needle probe method and design. Table I shows data acquired at room temperature for materials of various conductivity values as well as values for each material reported in the literature. Results show that the probe is able to measure thermal conductivity within the range of 0.2-16 W/m·K with an error range of less than 5% at room temperature.

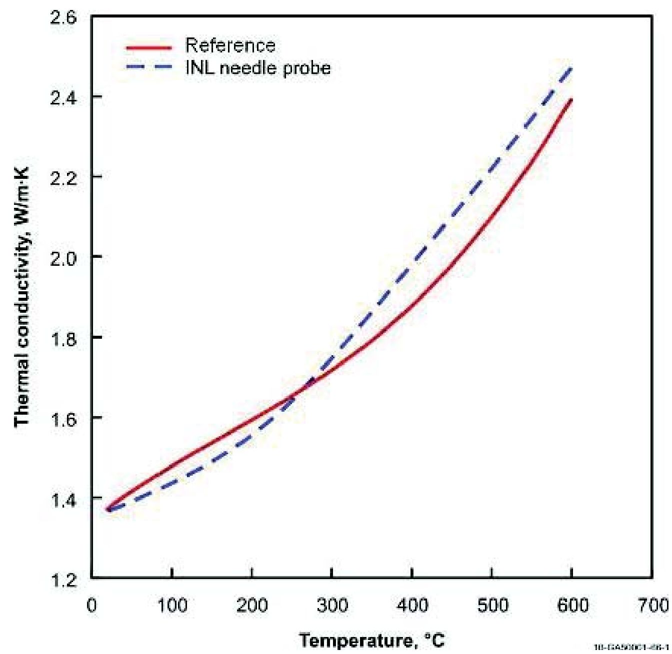

Fig. 8. Temperature dependant thermal conductivity of fused silica.

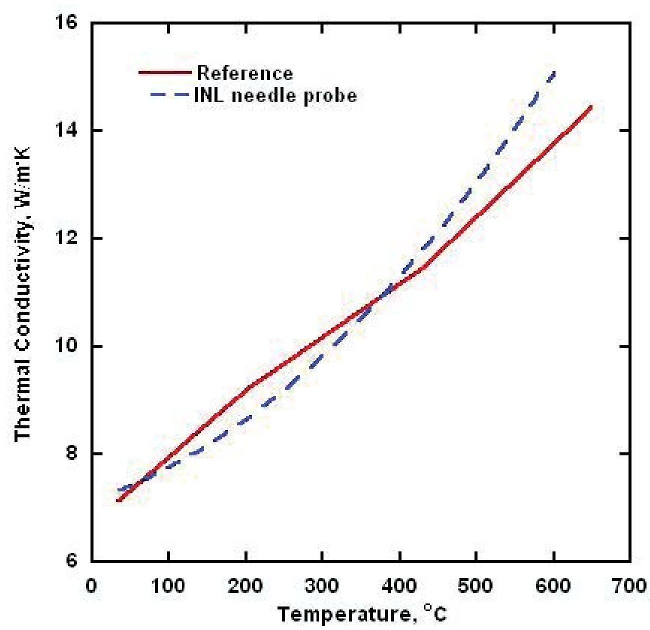

Fig. 9. Temperature dependant thermal conductivity of 6–4-titanium.

Thermal conductivity values for fused silica have also been acquired as a function of temperature and are shown in Figure 8. The needle probe data varies from the Touloukian data [10] by a maximum of 5.7% over the range of 20 to 600 °C (1.4 to 2.4 W/m·K).

Data collected for 6%Al-4%V-Titanium are shown in Figure 9. INL data vary from reference data [11] by a maximum of 7.7% over the range of 20 to 600 °C (7 to 15 W/m·K).

Data collected for Inconel 625 are shown in Figure10. INL data vary from reference data [12] by a maximum of 7.0% over the range of 20 to 600 °C (10 to 19 W/m·K).

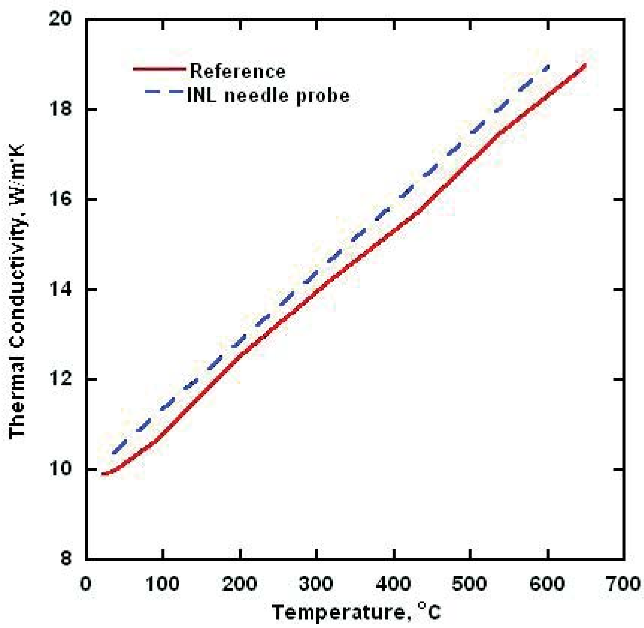

Fig. 10. Temperature dependant thermal conductivity of Inconel 625.

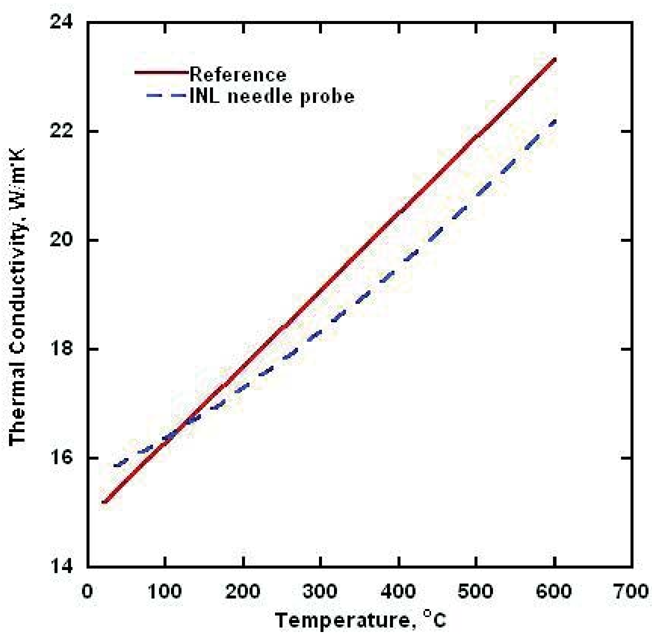

Fig. 11. Temperature dependant thermal conductivity of 304-stainless steel.

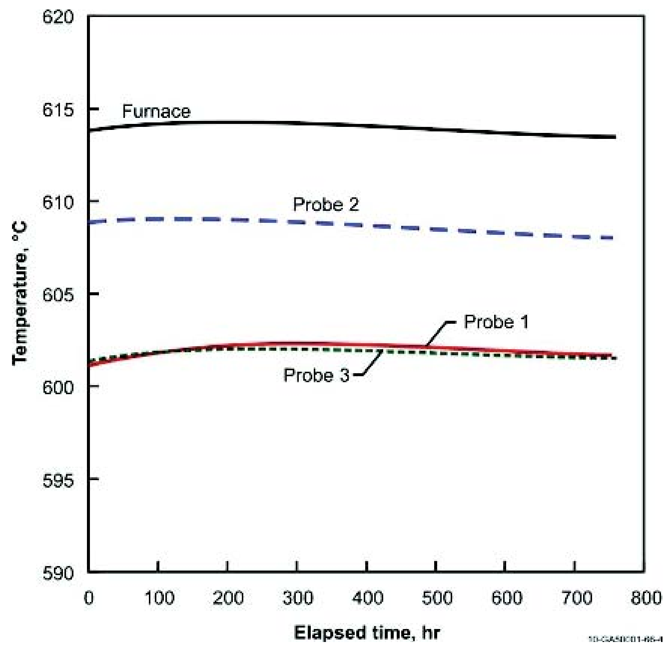

Fig. 12. Temperature data collected during long duration testing.

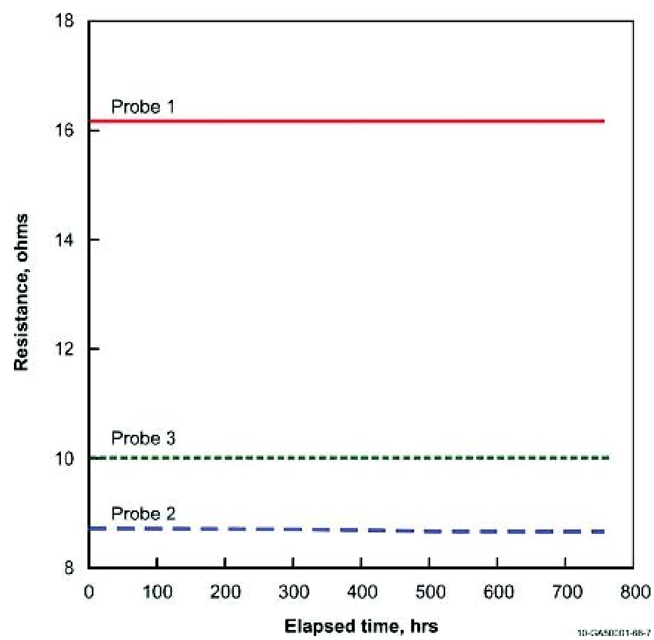

Fig. 13. Resistance values of heating elements in long duration test.

Data collected for 304-stainless steel are shown in Figure 11. INL data vary from reference data [10] by a maximum of 10.0% over the range of 20 to 600 °C (16 to 22 W/m·K).

A long duration test has been completed to assess the robustness of the needle probe. In this test, three needle probes were heated at 600 °C for over 1000 hours. The heating elements were energized with 1.5 volts for several minutes daily to simulate use in a test. Both temperature response of the thermocouples and resistance of the heating elements were monitored. Figure 12 shows temperature data for the three tested probes as well as the test furnace temperature. Results show that the design is robust yielding consistent temperature readings. Resistance values shown in Figure 13 for the heating elements are also very stable, indicating consistent power input (note that differences in resistance values are due to different diameter wires used in the probes, as each probe is slightly different in design).

III. Enhancement of Data Processing Methods

The ASTM standard for thermal conductivity measurement by the needle probe method [1] recommends measuring the temperature response in increments of 5 seconds or greater for a minimum of 1000 seconds. Thermal conductivity is then calculated using the average linear slope of two sets of data, one for heating the sample and one for cooling (both

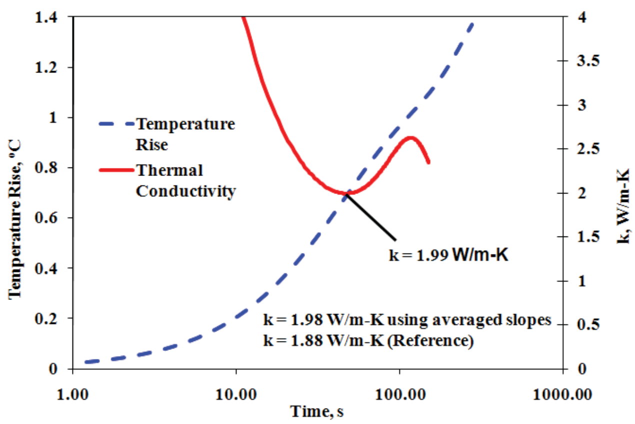

Fig. 14. Comparison of thermal conductivity values yielded by different data processing methods for fused silica at 400 °C.

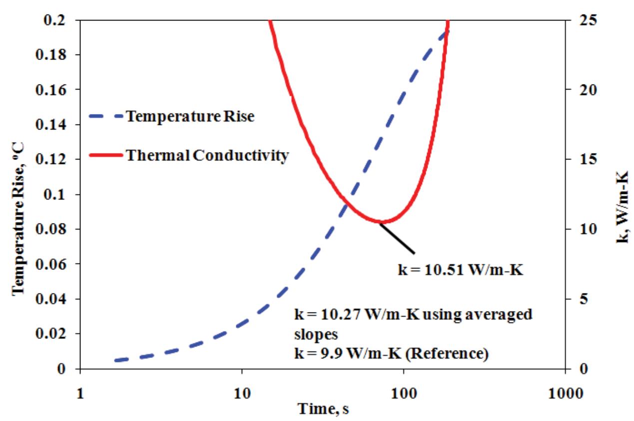

Fig. 15. Comparison of thermal conductivity values yielded by different data processing methods for Inconel 625 at 400 °C.

starting at steady state). This process is based on the typical use of the needle probe design, low temperature (<100 °C) measurement of soils, soft rocks, and food products with thermal conductivity values less than 5 W/m·K, and sample diameters of at least 50 mm. For higher thermal conductivity samples and smaller sample sizes, for which the linear region may be very short, the derivative of the temperature response with respect to the natural logarithm of time may be used in place of the average slope of the linear region [16]. This is important for laboratory work as it eliminates the need to use cooling data, and reaching steady state prior to cooling may take a prohibitively long time. Examples of this derivative based method and comparisons to the ASTM Standard method are shown in Figures 14 and 15.

IV. Conclusion

An effort has been initiated to develop and assess the performance of an in-pile sensor, based on needle probe techniques, for detecting thermal conductivity. To date, results from evaluations of a proposed needle probe design are very promising The needle probe has been demonstrated to work very well for materials with thermal conductivity ranges from 0.2 to 22 W/m·K with measurement errors of less than 5% for samples tested at room temperature, 6% for low thermal conductivity samples tested at high temperatures, and 10% for high thermal conductivity samples tested at high temperatures, delivering thermal conductivity measurements with a high degree of accuracy. The test results indicated that special design considerations are needed for materials with a high thermal conductivity, or samples with a small diameter. High thermal conductivity and small diameter sample materials pose some challenges, but methods for improving the technique are being developed, primarily to reduce signal noise and better characterization of the probe response time. Results from long term evaluations indicate that the needle probe is a robust sensor that could survive in harsh environments, such as measuring fuel conductivity in-pile. Future evaluations will include in-pile fuel conductivity measurement in the Massachusetts Institute of Technology Test Reactor, where small diameter high conductivity hydride fuel will be evaluated.

Acknowledgment

References herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the U.S. Government, any agency thereof, or any company affiliated with the Idaho National Laboratory, Idaho Falls, ID.

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She was a fellow of the American Nuclear Society in 2005. In 2009, she was elected to the ANS Board of Directors. She was selected to serve on the U.S. Nuclear Regulatory Commission's Advisory Committee on Reactor Safeguards in 2010. She received the U.S. Department of Energy Secretarial Honors Award in 2011. She serves as a member of several advisory groups reviewing the U.S. Department of Energy's Office of Nuclear Energy Research and Development programs and she provides consulting assistance to vendors, utilities, and regulators on severe accident issues.

Darrell L. Knudson has more than 30 years of experience in nuclear reactor safety analysis, fuel performance modeling, computer code development, and high-temperature instrumentation design, fabrication, and testing. He is currently leading High Temperature Test Laboratory (HTTL) efforts to develop the capability for in-core measurement of elongation using linear variable displacement transducers and ultrasonic techniques.