5-2010

In-Pile Thermal Conductivity Measurement Methods for Nuclear Fuels

Brandon S. Fox

Utah State University

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ABSTRACT

In-Pile Thermal Conductivity Measurement

Methods for Nuclear Fuels

by

Brandon S. Fox, Master of Science

Utah State University, 2010

Major Professor: Dr. Heng Ban
Department: Mechanical and Aerospace Engineering

Measuring nuclear fuel thermal conductivity in-pile can provide much needed data for understanding fuel performance during irradiation and yield thermophysical property data needed for simulation codes and fuel databases. The objective of this research is to develop and compare two in-pile thermal conductivity methods in a laboratory setting using surrogate fuel materials.

A steady-state radial heat flow method was investigated to understand its viability as an in-pile steady-state thermal conductivity technique. By using Joule heating to simulate volumetric heat generation within a surrogate fuel rod, thermal conductivity was measured with two thermocouples at different radial positions within the rod. Examinations were completed on two batches of surrogate materials over the temperature range of 500 to 700 °C. The selected surrogate rod was fabricated from the only material identified to possess the required thermal conductivity and electrical resistivity required for the selected labora-
tory approach. Evaluations estimated a measurement uncertainty of 12% and values were within 33% of values obtained using laboratory material property measurement systems for this surrogate material. Results indicate that the selected surrogate rod material limited the ability to assess this approach at higher temperatures in a laboratory setting.

A transient needle probe method adapted from American Standard Test Method standards was also used to measure temperature-dependent thermal conductivity of surrogate fuel rod materials for temperatures ranging from room temperature to 400 °C. The needle probe has a heating element and a temperature sensor contained in a metal sheath, and it is inserted into the surrogate fuel rod whose thermal conductivity is to be measured. The thermal conductivity is calculated from the power applied to the heating element, and the temperature rise detected in the sample. Needle probes were designed and fabricated using materials recommended for in-pile application. Scoping room-temperature values obtained using the needle probe method were within acceptable accuracies defined by the ASTM needle probe reference standard. Temperature-dependent values were within 2% of values for the well-characterized ASTM recommended reference material, fused silica. A measurement uncertainty under 6% was calculated for the needle probe method.

As a result of this study, the needle probe method was selected for additional testing at the Idaho National Laboratory for anticipated testing in Materials Test Reactors. This would result in the first-ever transient in-pile thermal conductivity sensor.
ACKNOWLEDGMENTS

I would like to express my thanks to the researchers at Idaho National Laboratory’s High Test Temperature Laboratory for their support, expertise, and direction in completion of this work. I would also like to thank my committee members and major professor for their time and effort on my behalf. Finally, I would like to express my gratitude to a loving wife, supporting family, caring friends, and three very active little boys who give me inspiration.

Brandon S. Fox
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C  Needle probe calibration constant

C_I  Fourier number factor from hot-wire references

C_p  Specific heat (J/kg°C)

Ei(x)  First order exponential integral function

I  Current (amps)

L  Length (m)

L/d  Ratio of probe length to outside diameter

L_o  Initial length (m)

P  Power (W)

P_γ  Gamma heating (W/cm³)

Q  Heat input per unit length (W/m)

R  Electrical resistance (Ohm)

S  Linear slope of temperature versus natural logarithm of time (°C)

T  Temperature (°C)

T_c  Centerline temperature (°C)

T_s  Surface temperature (°C)

T_{r1}  Temperature at probe center (°C)

T_{t1}  Temperature at time location 1 (°C)

T_{r2}  Temperature probe radius (°C)

T_{t2}  Temperature at time location 2 (°C)

V_o  Initial volume (m³)

α  Thermal diffusivity (m²/s)

α_L  Linear coefficient of thermal expansion (1/°C)
\( \alpha_1 \)  
Probe thermal diffusivity (m\(^2\)/s)

\( \alpha_2 \)  
Sample thermal diffusivity (m\(^2\)/s)

\( \gamma \)  
Euler–Mascheroni constant

\( \varepsilon \)  
Ratio of \( k_1 \) to \( k_2 \)

\( \varepsilon_i \)  
Current measurement uncertainty

\( \varepsilon_k \)  
Total measurement uncertainty

\( \varepsilon_L \)  
Length measurement uncertainty

\( \varepsilon_r \)  
Radial distance measurement uncertainty

\( \varepsilon_{ro} \)  
Rod radius measurement uncertainty

\( \varepsilon_S \)  
Slope calculation uncertainty

\( \varepsilon_V \)  
Voltage measurement uncertainty

\( \varepsilon_{AT} \)  
Temperature difference measurement uncertainty

\( \eta \)  
Ratio of \( (k_1/\alpha_1) \) to \( (k_2/\alpha_2) \)

\( \lambda \)  
Probe length to outside diameter ratio

\( \rho \)  
Density (kg/m\(^3\))

\( \rho_{electrical} \)  
Static electrical resistivity (Ohm-m)

\( \rho_f \)  
Final density (kg/m\(^3\))

\( \sigma \)  
Needle probe axial error factor from Blackwell [38]

\( \Delta L \)  
Change in length (m)

\( \Delta R \)  
Maximum relative error

\( \Delta T \)  
Temperature difference (°C)

\( \Delta V \)  
Change in volume (m\(^3\))
CHAPTER 1
INTRODUCTION

In-pile instruments are used to detect and measure various physical parameters of fuels and materials during irradiation [1]. In the case of nuclear fuels during irradiation, the physical structure and chemical composition change as a function of time and position within the fuel pellet. For example, fuel pellets can swell, crack (micro-cracking), and fission gases can be released. These conditions can vary with time in the reactor, temperature, and fuel burnup [2]. Hence, interpreting data from in-pile instrumentation can be extremely complicated.

Thermal conductivity is a key property of interest for both nuclear fuel and structural materials, and must be known for proper design, test, and application of new fuels and structural materials in nuclear reactors. Thermal conductivity is a transport property and is highly dependent on the physical structure, chemical composition, and the state of the material [3]. Typically, thermal conductivity changes that occur during irradiation are measured out-of-pile by Post Irradiated Examination (PIE) using a “cook and look” approach in hot-cells. Repeatedly removing samples from a test reactor to make out-of-pile measurements 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. 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 nuclear fuel data, reduce costs
from PIE examinations, increase understanding of how fuels behave under irradiation, and confirm or improve existing thermal conductivity measurement techniques.

Since the 1960s, researchers have tried to measure fuel thermal conductivity in-pile [4]. However, only one known in-pile nuclear fuel thermal conductivity technique is currently used at Materials and Test Reactors (MTRs), and this approach invokes several assumptions about the fuel composition and heat transfer within the fuel and its cladding [5].

The objective of this research is to investigate potential techniques to calculate and monitor fuel thermal conductivity in-pile. Two methods were investigated, a steady-state radial heat flow method using two thermocouples, and a transient method using a hot wire adaptation from American Society for Testing and Materials (ASTM) standards. This thesis identifies method limitations with respect to laboratory settings and reactor settings and recommends a technique for further in-pile consideration and implementation.

The first method calculates fuel rod thermal conductivity by applying Joule heating to simulate volumetric heat generation and using two thermocouples inserted into the surrogate fuel rod, one to monitor fuel centerline temperature and another to monitor temperature at a measured radial position within the rod.

The second method is based on Transient Hot Wire Methods (THWM). The selected needle probe method is based on the theory of an infinite line heat source in an infinite solid. The probe contains a heat source element and a temperature sensor inserted into a material whose thermal conductivity is to be measured. The surrogate nuclear fuel rod thermal conductivity is determined from the temperature rise measurement of the sample.
Preliminary investigations by researchers at the Idaho National Laboratory (INL) indicate that this approach may offer advantages over steady-state techniques [6].

This thesis is divided into eight chapters. Chapter 2 contains the literature review of methods and research experience related to the current work, as well as known method limitations from literature sources. Chapter 3 lists research objectives for both steady-state and transient method methods. Chapter 4 describes the methodology and procedure for the USU/INL steady-state method, which includes several important sub-sections. The method background, governing equations, and known practical limitations are detailed, as well as descriptions of the experimental setup, experimental procedure, measurement sensitivities examinations, and surrogate materials used for evaluating of the steady-state method. Similarly, Chapter 5 explains the methodology and procedure for the transient method, which includes several important sub-sections. The method background, governing equations, and known practical limitations are detailed, as well as a description of the experimental setup, experimental procedure, measurement sensitivities examinations, and surrogate materials to be used to evaluate the transient method. Steady-state two-thermocouple method results and discussions are given in Chapter 6, including surrogate material thermophysical properties measurement results for estimates of thermal conductivity, comparisons of two-thermocouple method experimental results to properties measured estimates of thermal conductivity, and measurement sensitivities to constant power, ambient temperature, and surrogate batch variations. Chapter 7 provides needle probe transient method room temperature and elevated temperature surrogate material results. Measurement sensitivities discussions are also given with respect to constant heater power setting, surrogate material measurements, ambient temperature, contact resistance between probe
sheath and surrogate fuel rod, and linear region slope calculations. Conclusions from both methods investigated in this study are detailed in Chapter 8, as well as suggestions for further consideration and study. References follow Chapter 8 and are listed in order of placement in text. The Appendix gives plots of all relevant data used in calculations from the needle probe method.
This chapter contains three major sections. The first section details anticipated reactor test conditions, a description of in-pile thermal conductivity measurement difficulties, and a description of the problem motivating this research. The second and third sections in the chapter provide a literature review of specific thermal conductivity measurements for in-pile applications. This review focuses upon two specific topics: steady-state and transient methods. Both sections identify previous research experience from other relevant studies, background of each method, and known identified method limitations. Because the focus of this research is directed toward in-pile measurements, only methods relevant to this research are considered. For a more detailed survey of general thermal conductivity measurements, see References [7] through [9].

2.1. Anticipated Testing Conditions

The ultimate goal from laboratory testing is to provide an in-pile thermal conductivity technique suitable to implement at INL’s Advanced Test Reactor (ATR) and other MTRs in the world.

In-pile testing presents numerous complications for instrumentation. For example, transmutation of sensor materials with large thermal neutron absorption cross sections can result in sensor decalibration during irradiation, as noted by Rempe et al. [10]. While in the reactor, the material under irradiation also experiences changes. For instance, the fuel densifies, swells, and cracks; gas composition and pressure changes within the cladding; and pellet-to-cladding interactions may occur.
In addition, high temperatures and large thermal gradients present additional sensor design and material selection complications. For example, the temperature gradients within the fuel can be large, and the gradient from the fuel centerline to the cladding surface is also large. An example of this gradient is seen in Table 2-1 for typical commercial Pressurized Water Reactor (PWR) conditions, where the difference between the maximum fuel center temperature and cladding surface temperature is 1535 °C. Understanding these temperatures and gradients is critical to in-pile thermal conductivity sensor design.

Table 2-1. Typical Commercial Reactor Fuel and Cladding Parameters [11]

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Reactor Type</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>PWR</td>
</tr>
<tr>
<td>Fuel Material</td>
<td>UO₂</td>
</tr>
<tr>
<td>Pellet Height</td>
<td>0.6 in (1.5 cm)</td>
</tr>
<tr>
<td>Pellet Diameter</td>
<td>0.37 in (0.9 cm)</td>
</tr>
<tr>
<td>Maximum Fuel Center Temperature</td>
<td>3420 °F (1882 °C)a</td>
</tr>
<tr>
<td>Maximum Linear Heat Rateb</td>
<td>42.7 kW/m</td>
</tr>
<tr>
<td>Average Linear Heat Rateb</td>
<td>17.8 kW/m</td>
</tr>
<tr>
<td>Cladding</td>
<td>Zircaloy-4</td>
</tr>
<tr>
<td>Material</td>
<td></td>
</tr>
<tr>
<td>Outer Diameter</td>
<td>0.422 in (1.07 cm)</td>
</tr>
<tr>
<td>Thickness</td>
<td>0.024 in (0.06 cm)</td>
</tr>
<tr>
<td>Average Temperature</td>
<td>657 °F (347 °C)</td>
</tr>
</tbody>
</table>

a. This temperature was taken from Rust [12].
b. Refer the reader to Todreas and Kazimi [13].
Fuel pellet and cladding geometries are also important factors for in-pile thermal conductivity sensors. Measurement sensors must be able to accommodate small fuel pellet volumes, as seen in typical Boiling Water Reactor (BWR) fuel pellet designs from Table 2-1.

Testing in the ATR can vary depending on several factors. Thus, thermal conductivity measurement sensors must be adaptable to desired test conditions. In fuel measurements, hole/s may be drilled in the fuel to accommodate sensors. The fuel is loaded into a test capsule and positioned in the reactor with instrument leads extending from the reactor to the data acquisition systems. The thermal conductivity measurement is subject to specific geometries inside the instrumented lead experiment capsule. Thus, applicable in-situ thermal conductivity measurement techniques must accommodate radiation effects, high temperatures, large thermal gradients, and geometry constraints of the actual material sizes being evaluated within the reactor (e.g., limited irradiation test volumes).

2.2. Steady-state Radial Heat Flow Methods

Steady-state methods to measure thermal conductivity depend largely on the conduction rate equation, Fourier’s law, which is defined in heat flux form by Equation (2.1).

\[ q'' = -k \frac{dT}{dx} \]

where:

- \( q'' \) = heat per unit area (W/m\(^2\)),
- \( k \) = thermal conductivity (W/mºC),
- \( T \) = temperature (ºC), and
- \( x \) = conduction distance (m).
Equation (2.1) implies that heat flux has directionality, meaning the heat flux will always be normal to the surface of constant temperature [14]. In cylindrical systems with internal heat generation, the heat conduction in a solid, homogenous, infinitely long bar will flow radially from the center to the surface. A temperature distribution is created in the bar, and at steady-state conditions, the temperature distribution does not change with time. Equation (2.1) can be rearranged to calculate thermal conductivity when the temperature difference, heat flux, and cylindrical geometries are known.

Most radial heat flow methods typically use a cylindrical core heater surrounded by an outer heater, where the sample is in the annulus between the heaters. However, variations in this method are common. The heaters supply a temperature difference in the rod so that a form of Equation (2.1) can be used to calculate thermal conductivity.

Radial heat flow methods have commonly been used to measure thermal conductivity of various materials at various temperatures (e.g., see References [9] and [15]). Simplicity is the largest advantage of radial flow methods compared to other steady-state and transient methods. Waiting for steady-state equilibrium can be a disadvantage with all steady-state methods, as a result, time periods between data points range between hours and days or weeks depending on the specific method and material.

2.2.1. In-Pile Steady-State Thermal Conductivity Research
Experience from Cylindrical Radial Heat Flow Methods

As mentioned, there are few techniques available for in-pile thermal conductivity detection; consequentially, there are few reference sources on this subject. This is mainly due to two facts: first, other than nuclear fuel elements, there are few applications of heat generation, and second, in-pile instrumentation, specifically thermal conductivity mea-
surements, have numerous complications and are costly. Cohen et al. [4] detail efforts as early as 1960, while the Institute for Energy Technology at the Halden Reactor Project (IFE/HRP) researchers (see References [1], [2], and [5]) describe the only known current method in use today. These efforts describe similar techniques using the well-known heat transfer principle of radial heat flow applied to a general two-thermocouple approach. Table 2-2 summarizes key attributes from these known research efforts.

2.2.1.1. Bettis Atomic Power Laboratory Experience

Cohen et al. [4] describe the calculation of Uranium Dioxide (UO₂) thermal conductivity in-pile made by a two-thermocouple method, where a Pt-10 Rh centerline thermocouple inserted in a hole drilled through seven and one-half fuel pellets is used with the inner cladding temperature to back-calculate the “effective” thermal conductivity, as seen in Figure 2-1.

Table 2-2. Key Attributes of In-Pile Thermal Conductivity Research Experiments

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Fuel</td>
<td>Cold-pressed and sintered UO₂</td>
<td>Uranium-5 wt% fissiuma</td>
<td>Various fuel types</td>
</tr>
<tr>
<td>Sensor type</td>
<td>Pt-10% Rh / Fe-Constantan Thermocouples</td>
<td>“TC” (type not defined)</td>
<td>Type C thermocouple</td>
</tr>
<tr>
<td>Sensor diameter/s</td>
<td>0.159 / 0.165 cm</td>
<td>NA</td>
<td>0.158 cm</td>
</tr>
<tr>
<td>Location of sensor/s</td>
<td>Centerline / cladding</td>
<td>Fuel and coolant</td>
<td>Centerline only</td>
</tr>
<tr>
<td>Test temperature range</td>
<td>0-1200 ºC</td>
<td>NA</td>
<td>0-1500 ºC</td>
</tr>
</tbody>
</table>

a: The major constituents of fissium are zirconium, molybdenum, palladium, ruthenium, and rhodium. Fissium is an equilibrium concentration of fission product elements from the Experimental Breeder Reactor-II (EBR-II) pyrometallurgical reprocessing cycle [16].
This experiment measures the gamma heating along with the fuel thermocouple response, which is then used along with sample geometry to calculate the heat generation rate from Equation (2.2).

\[
k \Delta T = \frac{q'}{2\pi} \ln\left(\frac{r_o}{r_i}\right) + \frac{P_\gamma}{2} \left[ \frac{r_o^2 - r_i^2}{2} - r_i^2 \ln\left(\frac{r_o}{r_i}\right) \right],
\]

(2.2)

where:
- \( k \) = thermal conductivity of 304 stainless steel (SS) (W/cm°C),
- \( q' \) = heat generation rate per unit length in oxide (W/cm),
- \( r_o \) = outer radius (cm),
- \( r_i \) = inner radius (cm),
- \( \Delta T \) = temperature difference (°C) between \( r_i \) and \( r_o \), and
- \( P_\gamma \) = gamma heating (W/cm³).
The effective thermal conductivity can then be evaluated using,

\[
\int_{r_1}^{r_o} k \, dk \, \theta = k' \Delta T = \frac{\dot{q}}{2} \left[ \frac{r_o^2 - r_1^2}{r_1} - r_1^2 \ln \left( \frac{r_o}{r_1} \right) \right],
\]

(2.3)

where:

- \( k \) = UO₂ thermal conductivity (a function of temperature) (W/m\( ^\circ \)C),
- \( k' \) = “effective” thermal conductivity (W/cm\( ^\circ \)C),
- \( \dot{q} \) = heat generation rate (W/cm\(^3\)) = \( q'' \left[ \frac{1}{\pi (r_o^2 - r_1^2)} \right] \),
- \( r_o \) = outer radius of fuel (cm),
- \( r_1 \) = inner radius of thermocouple well (cm),
- \( \Delta T \) = temperature difference between SS bore and fuel center (°C), as seen in Figure 2-1,
- \( T_c \) = centerline temperature of fuel (°C), and
- \( T_s \) = surface temperature of fuel (°C).

Using Equations (2.2) and (2.3), Cohen et al. were able to generate plots of thermal conductivity as a function of temperature and rod burnup for several variations of UO₂ fuel and chamber fluids. The temperature range was 75 - 1175 °C, and the results were correlated with only a few out-of-pile references. In fact, the authors indicate their in-pile measurements did not always correspond with PIE measurements.

There are acknowledged complications by Cohen et al. about quantifying the heat transfer in the fuel elements. The first is the sharp change in thermal conductivity of UO₂ from already very low values, making accurate measurements difficult. The second is the diametrical clearance between the fuel rod and the cladding. Estimating the effects on gap conductance and fuel performance as the gap between the fuel and cladding changes is difficult because brittle ceramic fuel cracks as it swells during irradiation. Cohen et al. also substantiated that out-of-pile measurements did not explain structural changes of irradiated UO₂ from PWR type rods under several heat generation conditions. It also reports
that out-of-pile thermal conductivity of sintered UO$_2$ from numerous sources differ as much as 35%.

2.2.1.2. Argonne National Laboratory Experience

Another two-thermocouple method to detect thermal conductivity was investigated at the Experimental Breeder Reactor II (EBR-II) at Argonne National Laboratory (ANL), where experimental thermocouple data from both fuel and coolant was “augmented by other analytical calculations” to back-calculate the thermal conductivity of Uranium-5 wt% fissium [16]. Three important regions of thermal conductivity as a function of rod burnup are identified from the ANL research, as seen in Figure 2-2. Burnup is a measure of fuel atoms undergoing fission and effectively identifies the energy potential in the fuel. Figure 2-2 is adapted from Betten [16] to show the measured thermal conductivity normalized by the beginning of life value, $K_o$, for a single element, and illustrates fuel burnup has a large impact on fuel thermal conductivity. Figure 2-2 identifies three regions of interaction between the cladding and fuel. Because fuel swelling affects gap clearance and gap conditions are needed to calculate or measure the heat transfer from the fuel to the coolant, it is very important to understand how the gap changes with burnup. Fuel swells in both radial and axial directions, and region 1 represents the as-assembled clearance between the fuel and cladding where there is no interaction. As the fuel begins to swell and contacts the cladding, fission gas is released and can be monitored, as reported by Wiesenack and Tverberg [1]. Betten also speculates that region 2 temperature peaks are from the first interactions between fuel/cladding and fission gas release, as seen in Figure 2-2. Region 3 is over a wide range of atomic% of burnup; and as speculated by Betten,
represents the physical condition of the fuel in contact with the cladding and the equilibrium interconnected porosity condition.

Results from Betten also identify the difficulty of measuring thermal conductivity in-pile because of reactor conditions. Accommodations for fuel swelling must be made when in-pile thermal conductivity techniques are employed, and in many cases these accommodations are difficult to predict. For example, with brittle ceramic fuels where swelling is accompanied with micro-cracking [1].

2.2.1.3. Halden Reactor Project Experience

Currently, the Halden Boiling Water Reactor (HBWR) is the only test reactor where in-pile fuel thermal conductivity measurements are performed (References [1], [2], and [5]). IFE-HRP researchers use this technique to assess the impact of burnup on thermal
conductivity, where a fuel centerline thermocouple, well-known heat flux and thermal hydraulic conditions are used to calculate the thermal conductivity as a function of temperature and fuel burnup, as seen in Figure 2-3. This approach, along with other in-pile approaches, must assume several conditions about the fuel, 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 center temperature during irradiation. Hence, the approach requires non-prototypic fuel rods and is susceptible to uncertainties associated with the assumptions that must be invoked.

Figure 2-3. HRP measured UO₂ thermal conductivity as a function of temperature and burnup [5].
2.2.2. Steady-State Radial Heat Flow In-Pile

Thermal Conductivity Method Summary

The largest limitation from radial heat flow methods is the required time to reach steady-state, which in some cases can be many hours for only one data point. Presley and Christensen [9] offer laboratory measurement accuracies less than 4% for the radial heat flow method with causes of inaccuracies seen from axial heat loss errors, radiation losses, thermal expansion of the sample, unsymmetrical heat flow, and poor thermal contact between the sensor and sample. Most of these causes of error can also be large in-pile steady-state method contributors of measurement error.

2.3. Transient In-Pile Methods

There exists no known transient in-pile thermal conductivity measurements method. Because of the measurement requirements listed in Section 2.1, only specific transient methods can be applied for in-pile measurements. Therefore, potential transient methods are discussed in this section. In particular, the line heat source method, and the thermal conductivity probe or needle probe method.

2.3.1. Thermal Conductivity Probe

The line heat source theory was suggested by Schleiermacher [17] and later by Stalhane and Pyk [18]. One of the first practical uses of the method measured the thermal conductivity of liquids by Van der Held and Van Drunen [19]. The mathematical approach was suggested by Carslaw and Jaeger [20] using the assumption of radial diffusion of heat from a line source, as seen in Equation (2.4) from the details Presley and Christensen [9].

\[
\frac{\partial T}{\partial t} = \frac{\alpha}{r} \frac{\partial}{\partial r} \left( r \frac{\partial T}{\partial r} \right),
\]  

(2.4)
where:

\[ \alpha = \frac{k}{\rho C_p}, \]
\[ k = \text{sample thermal conductivity (W/m}^\circ\text{C)}, \]
\[ \rho = \text{sample bulk density (kg/m}^3\text{)}, \]
\[ C_p = \text{specific heat of the sample (J/kg}^\circ\text{C)}, \]
\[ T = \text{temperature (}^\circ\text{C)}, \]
\[ t = \text{time (s), and} \]
\[ r = \text{is the radial distance from the heat source (m)}. \]

Equation (2.4) can be solved for the transient temperature response, \( T(r,t) \), by the thermal equilibrium initial condition, where the temperature rise in the sample, \( T(r,0) = 0 \). The two required boundary conditions are: (1) the far-field boundary condition, where the temperature rise in the sample, \( T(\infty,t) = 0 \); and (2) the line heater surface heat flux condition, as seen in Equation (2.5),

\[
-2\pi r k \left( \frac{\partial T}{\partial r} \right) = Q',
\]

where, \( Q' \) is the power per unit length (W/m) of the heat source after time zero.

Numerous references may be found in the literature describing applications of this method to measure the thermal conductivity of solids, fluids, and gases (e.g., see References [21] through [28]). This technique has been accepted as a standard method to calculate the thermal conductivity of materials ranging from thermal insulators [24] with conductivity values of 0.02 W/m\( ^\circ \text{C} \) to rock samples with conductivity values around 10 W/m\( ^\circ \text{C} \) [29]. Today, several vendors offer systems measuring thermal conductivity based on THWM techniques (e.g., see References [27] and [30]).

In a solid, this method is applied by embedding a line heat source (can be either embedded wire or heating element contained in a sheath) in the material whose thermal conductivity is to be measured, as illustrated in Figure 2-4 (top). From a condition of ther-
mal equilibrium, the heat source is energized and heats the medium with constant power. The temperature sensor is located at a known radial distance from the heat source, and the temperature response in the medium from the sensor is a function of its thermal properties. The thermal conductivity is calculated from the temperature rise detected in the sample.

The thermal conductivity probe or needle probe method theory is based on the line heat source theory. This is also a well-known and often used method for measuring thermal conductivity of various materials and test conditions (e.g., see [31] for the ASTM Standard measurement of soils and rocks, [26] for porous and granular materials, [30] for food and agriculture, [32] for large probe designs, [33] for high temperature melts, and [34] for measuring thermal diffusivity and specific heat from thermal conductivity probe data).

Figure 2-4. Illustration of the THMW or line heat source method (top), and needle probe method (bottom).
The major advantage of the needle probe method is the potential for *in-situ* measurements, as seen in the illustration of the needle probe from Figure 2-4 (bottom), where a single sensor is inserted into the material to be measured. Probes typically consist of a hollow tube housing a heat source element and a temperature sensor separated by a medium with good electrical insulation and high thermal conductivity to ensure negligible radial temperature differences within the probe. For measurement, samples are prepared with a hole machined to accommodate the probe outer diameter (e.g., for solids); or probes are simply pressed into the sample to be measured (e.g., for soils, liquids, or tissues). Impacting the test sample with only one, small hole is an advantage of the needle probe method over the line heat source method.

### 2.3.2. Previous INL Experience

Preliminary investigations by Rempe et al. [6] at INL's High Temperature Test Laboratory (HTTL) suggest that hot wire methods would be viable for measuring thermal conductivity of materials during irradiations in INL's ATR. Advantages of line heat source method are its potential to yield data with shorter measurement times (than possible with steady-state methods) and its ease of installation as an in-pile sensor (compared to other transient methods, such as, transient pulsed diffusivity methods used in specialized laboratory systems). INL explored the THWM in a laboratory setting, and lessons learned were incorporated into this effort.

### 2.3.3. Needle Probe Limitations

There are limitations to the needle probe method, primarily associated with experiment design and measurement practices. Because of the deviation from the line heat
source theory from practical applications, several factors need to be considered before applying the method to measure thermal conductivity. The summary given in this sub-section includes considerations to account for probe finite diameter, probe calibration, axial heat flow, finite sample size, heating rate, thermal contact, and linear region slope calculation.

2.3.3.1. *Probe Finite Diameter Time Correction*

Mohsenin [35] provides details to account for the finite radius of the line heat source. A time correction factor, $t_o$, is subtracted from the each observed time to allow for heat produced by the probe before the start of the measured time. The correction can minimize any resistance to heat transfer from the probe to the sample. Waite et al. [34] suggests a method to account for probe response time by comparing the time delay of two reference materials of different thermal conductivities. During the early transient for both reference materials, plots of temperature versus natural logarithm of time of the two materials are almost identical as heat builds up in the probe. Once the probe begins to transfer heat to the sample, thermal transfer is more efficient with the higher conductivity material; and the sample response is different than that of the lower conductivity material. An exact point can be seen where the sample response differs between the two reference materials, and the probe response time is known from this diverging point.

2.3.3.2. *Probe Calibration*

Some papers suggest that a small diameter probe design would eliminate the need for the time correction factor, and that a probe or calibration constant may help to account for the finite radius of the line heat source [35]. Mann and Forsyth [36] measured thermal
conductivity of insulation and insulation materials with various needle probes to identify the correlation of the calibration constants of each probe. They were able to calibrate the probes to account for deviations in raw data.

The calibration constant, $C$, as described in ASTM Standard D 5334-08 [31] depends primarily on the probe diameter. The ASTM Standard states that the calibration constant becomes less important with small probes. The thermal conductivity calibration constant must be applied for large diameter probes ($d > 2.5$ mm) where the error is greater. Hanson et al. [32] indicate that the calibration factor can also be a function of conductivity for large diameter probes. ASTM Standard D 5334-08 calls for testing with a calibration standard material with a well known thermal conductivity within the range of 0.2 - 5 W/m°C, such as: dry Ottawa sand, Pyrex 7740, Fused Silica (SiO$_2$), Pryoceram 9606, glycerine (glycerol). According to the ASTM Standard, the cylindrical calibration standard must be at least 20% longer than the probe length and 10 times the diameter of the probe diameter with a hole in the center to accommodate the exact probe length and diameter.

2.3.3.3. Axial Heat Flow Error

The ratio of the probe length to outside diameter, $L/d$, is an important value to indicate possible error from axial heat flow within the probe. The line heat source theory assumes one-dimensional radial heat flow, and small $L/d$ ratios may indicate more induced error from axial flow. Anter researchers [27] note that investigators have recommended minimum values for the heater $L/d$ ratios which range from 31 to 100. Hooper and Lepper [37] also recommend a $L/d$ ratio of at least 100 to minimize error from axial heat flow.

Blackwell [38] derived an expression for an upper limit to the axial flow error, as seen in Equation (2.6)
where:
\[ \Delta R = \text{maximum relative error}, \]
\[ \lambda = \text{length to outside diameter ratio}, \]
\[ \sigma = \text{four times the probe wall thickness divided by outside diameter for hollow probes, unity for solid probes}, \]
\[ \varepsilon = \text{ratio of thermal conductivity of probe, } k_1, \text{ to sample thermal conductivity, } k_2, \]
\[ \eta = \text{ratio of conductivity to diffusivity of the probe, } (k_1/\alpha_1), \text{ to the ratio of conductivity to diffusivity of the sample, } (k_2/\alpha_2). \]

Using Equation (2.6), Blackwell and Misener [39] showed that a probe with an outside diameter of 1.25 inch, a wall thickness of 0.125 inch, and \( L/d \) ratio of only 25, led to an error of less than 1% error.

2.3.3.4. Finite Sample Geometry Considerations

The line heat source theory assumes heat is transferred to an infinite medium (boundary condition (1) from Section 2.3.1); thus boundary edge effects are neglected. This infinite medium assumption is practically unachievable as sample geometries are finite. Thus, errors arise in the measurement if boundary temperatures experience a temperature change. Mohsenin [35] suggests shortening the measurement time and increasing sample diameter to minimize errors. Also, when possible, an additional temperature sensor at the sample boundaries can detect a temperature rise.

For accuracies of 1%, Prelovsek and Uran [24] recommend that the ratio of the sample diameter, \( d_{\text{sample}} \), to heater probe diameter, \( d_{\text{probe}} \), be at least 60 and that the length of the sample, \( L_{\text{sample}} \), be selected based on its thermal diffusivity and heating time, \( t_{\text{heating}} \), using the following relationship,
Analytically, a minimum sample diameter can be estimated by the expression $\frac{4\alpha t}{d^2}$, where $\frac{\alpha t}{d^2}$ is the Fourier number, a common dimensionless number with $\alpha$, being the thermal diffusivity (m$^2$/s), $t$, is the characteristic time (s), and $d$, is the shortest length through which conduction occurs in the sample (m). With respect to the sample diameter, References 26 - 28 and 40 suggest that the following be considered:

$$t_{\text{heating}} < C_1 \left( \frac{d_{\text{sample}}^2}{4\alpha} \right),$$

with $C_1$ varying from 0.1 (Van Gelder [28]), 0.15 (Anter Corporation [27]), 0.6 (Vos [40]) to 1 (Jones [26]) for accuracies of 1%. ASTM Standard D 5334-08 suggests “small diameter” probes to be less than 2.5 mm.

### 2.3.3.5. Heating Rate Effects

The accuracy of the measured thermal conductivity is enhanced if $t_{\text{heating}}$ is sufficiently large to clearly identify the linear region. For example, Manohar et al. [25] suggest the experimentalist vary the heater power level applied to a particular material using the following guidance:

- $t_{\text{heating}}$ be greater than 800 seconds for most materials
- early leveling of the temperature versus the natural logarithm of time plot for times less than 800 seconds be an indication that the power supply to the probe is too low
- a rapid increase in sample temperature is a sign that power to the probe is too high, causing heat build-up in the probe which can result in probe damage.
ASTM Standard D 5334-08 recommends that the temperature rise within the sample be no more than 10 ºC in 1000 seconds, so heater power selection must be considered. However, resolution of the slope of the temperature versus natural logarithm of time improves with increasing heater power.

2.3.3.6. Sample and Probe Thermal Contact

Good thermal contact between the probe and the sample material increases accuracy. Poor contact will delay the material response time, and thus, make slope estimates difficult. To enhance accuracy, thermal grease is recommended by ASTM Standard D 5334-08 to minimize uncertainties from poor sample-to-probe contact. Presley and Christensen [9] recommend using a highly conductive sheath to improve gap conductance, such as, stainless steel or aluminum and its alloys.

2.3.3.7. Data Collection and Slope Calculation

With power held constant, resistance heat is transferred from the probe to the sample and a plot of temperature versus the natural logarithm of time is created. From this data, the slope can be found and thermal conductivity of the sample can be calculated. ASTM Standard 5334-08 suggests that the first 15-30 seconds of the heating and/or cooling data be discarded so that the linear portion of the temperature versus the natural logarithm of time plot can clearly be seen. Manohar et al. [25] detail a properly configured test should identify three distinct segments. The first segment is the initial transient. The second is the linear segment where the slope should be clearly defined. The third segment is the final transient to steady-state condition and is dominated by boundary edge effects.
ASTM 5334-08 recommends using the average of the heating and cooling slopes when computing the thermal conductivity. The average of both heating and cooling slopes minimizes the temperature drift from multiple tests where heat build-up in the sample occurs, which can cause large uncertainties in calculations when only the heating slope is used for thermal conductivity calculations.

2.3.4. Transient Needle Probe Method Thermal Conductivity Measurement Summary

The needle probe method offers advantages over the steady-state approaches, mainly reduced measurement times from hours or days to only seconds or minutes. This approach minimizes temperature response measurements, and focuses more on material response from only one fuel centerline sensor and less on quantizing difficult parameters such as in-pile fuel to cladding contact resistance needed for radial heat flow two-thermocouple approaches.
CHAPTER 3
RESEARCH OBJECTIVES

This research investigates a steady-state and transient technique for in-pile nuclear fuel thermal conductivity measurement. Pre-experiment evaluations were conducted to examine potential test temperature ranges and surrogate materials for each method. Experiments were conducted at INL’s HTTL, and experimental results were presented at technical conferences and submitted for peer-reviewed journal publication.

3.1. Steady-State Method Research Objectives

The steady-state two-thermocouple method research objective is to assess its viability for detecting in-pile thermal conductivity by exploring the benefits and limitations in a laboratory setting, specifically by the listed objectives.

• Design and assemble components required for experimental setup to simulate volumetric heat generation by Joule heating in the selected surrogate material.
• Because the thermal conductivity of selected surrogate material is not well known as a function of temperature, temperature-dependent thermal conductivity measurements are made using standard laboratory material property measurement systems (e.g., pushrod dilatometer for estimates of density from thermal elongation measurement, differential scanning calorimeter for estimates of specific heat capacity, and laser flash for estimates of thermal diffusivity).
• Experimentally measure the surrogate material by the two-thermocouple method in a tube furnace over the temperature range of 500-700 °C.
• Quantify accuracy of the technique by comparisons of two-thermocouple experimental measured thermal conductivity results with estimated thermal conductivity values from material property measurements.

• Estimate the experimental measurement uncertainty range.

• Investigate steady-state method measurement sensitivities with respect to constant supplied power setting, controlled ambient temperature from tube furnace setpoint setting, and measurement variations from two batches of selected surrogate material.

• Suggest considerations and recommendations for additional study.

3.2. Transient Method Research Objectives

The transient needle probe method research objective is to explore the benefits and limitations of the needle probe method as an in-pile thermal conductivity measurement technique, specifically by the listed objectives.

• Design and fabricate needle probes for room temperature and temperature-dependent measurements from recommended materials for nuclear applications.

• Select surrogate materials to measure thermal conductivity for room temperature and elevated temperature testing based on anticipated nuclear fuel thermal conductivity values and ASTM recommended reference materials for needle probe measurements.

• Design and assemble experimental setup.
• Experimentally measure thermal conductivity by the proposed transient method for all selected surrogate materials at room temperature using both the room temperature designed needle probe and the high temperature designed needle probe.

• Experimentally measure thermal conductivity by the proposed needle probe technique in a tube furnace of the well-characterized ASTM recommended reference material, SiO$_2$, at three temperatures (e.g., room temperature, 250 °C, and 400 °C).

• Quantify accuracy of technique by room temperature and temperature-dependent comparisons of reported thermal conductivity values to experimentally measured thermal conductivity calculations.

• Estimate the experimental measurement uncertainty range.

• Investigate transient method measurement sensitivities with respect to varying parameters, such as surrogate material, constant power in the needle, controlled ambient temperature from tube furnace setpoint temperature, and thermal grease to enhance gap conductance.

• Suggest considerations and recommendations for additional study.
CHAPTER 4

STEADY-STATE METHOD APPROACH AND PROCEDURE

This chapter provides setup and procedure details for the steady-state method. The chapter includes theoretical background information and governing equations related to development and testing of thermal conductivity sensors. The surrogate material used for method evaluation and description of properties measurements needed to define the surrogate material temperature-dependent thermal conductivity are provided in this section.

4.1. Steady-State Two-Thermocouple Method

As discussed in Chapter 2, the two-thermocouple method is based on a well known heat transfer phenomenon [14], where heat generated within a rod flows radially to the surface. The temperature profile within the rod can be determined if rod geometry, material properties, and heat generation rate are known. The two-thermocouple method described in this research uses two thermocouples embedded in the rod to measure temperatures while volumetric heat generation is simulated by Joule heating from a measured input power source. Knowing two temperatures from different radial locations in the rod, power supplied to the rod, and rod geometry, the thermal conductivity of a material can be calculated. This approach deviates from previous in-pile steady-state methods because two thermocouples are embedded in the fuel, where others have used a centerline and a cladding thermocouple or the coolant temperature to calculate thermal conductivity.

4.1.1. Background

Incropera et al. [14] derive the steady-state temperature distribution in a solid, long rod
with uniform heat generation and uniform radial thermal conductivity. Physically, this derivation explains heat loss leaving the surface of a rod maintained at a constant value to be equal to the heat generated in the rod. Figure 4-1 shows the basis of this derivation,

where:

\[ \dot{q} = \text{volumetric heat generation rate (W/m}^3\text{)}, \]
\[ r = \text{radial position within the rod (m)}, \]
\[ r_o = \text{radius of the rod (m)}, \]
\[ L = \text{rod length (m)}. \]

4.1.2. Governing Equations

Equation (4.1) defines the cylindrical form of the heat conduction equation with constant, uniform, internal heat generation rate,

\[ \frac{1}{r} \frac{d}{dr} \left( r \frac{dT}{dr} \right) + \frac{\dot{q}}{k} = 0. \]  

(4.1)

The first boundary condition for this problem is of the first kind. For steady-state conditions, this represents the balance between heat generated in the sample and heat removed
by ambient conditions, as seen in Equation (4.2),

\[ T(r_o) = T_s, \quad (4.2) \]

where \( T_s \) is the surface temperature (°C). The second is a homogeneous boundary condition of the second kind, which physically represents a symmetry boundary condition, as seen in Equation (4.3)

\[ \left( \frac{dT}{dr} \right)_{r=0} = 0. \quad (4.3) \]

After applying boundary conditions, Equation (4.1) can be integrated to define the temperature distribution in the rod as a function of radial position, given by Equation (4.4),

\[ T(r) = \frac{\dot{q} r_o^2}{4k} \left( 1 - \frac{r^2}{r_o^2} \right) + T_s. \quad (4.4) \]

When \( r = r_o \), the temperature is the surface temperature, \( T_s \); and when \( r = 0 \), the temperature is the surface temperature plus the centerline temperature which is a function of the heat generation rate, rod geometry, and material thermal conductivity. The temperature can be defined at any location within the rod from Equation (4.4).

Equation (4.4) can be used to estimate the temperature at the fuel rod centerline, \( T(0) \),

\[ T(0) = \frac{\dot{q} r_o^2}{4k} + T_s. \quad (4.5) \]

Defining \( \Delta T = T(0) - T(r) \) to be the temperature difference between the centerline and the radial position, Equation (4.4) and Equation (4.5) can be combined to obtain the following relationship for thermal conductivity,

\[ k(r) = \frac{\dot{q} r^2}{4\Delta T}. \quad (4.6) \]
Hence, thermal conductivity of the two-thermocouple method can be calculated if the radial position from the sample centerline, \( r \); volumetric heat generation rate, \( \dot{q} \); and measured temperature difference, \( \Delta T \), are precisely known.

The derivation in Section 4.1.2 assumes constant thermal conductivity in the material; however, the thermal conductivity is temperature-dependent in most materials. If there exists a large temperature difference within a uniform material, the material thermal conductivity can vary with radial position. Material property measurements often assume the temperature difference in the material to be negligible, and a constant value can be assumed. For nuclear fuel elements, the temperature difference between the centerline and the cladding may be significant, and variations of thermal conductivity within the fuel rod can be large. For example, Cohen et al. [4] report measured temperature drops from the fuel centerline to the cladding surface of 240 °C, and Betten [16] reports 65 °C from the fuel centerline is assumed) to the coolant. Kakac and Yener [3] derive the temperature-dependent thermal conductivity for a cylindrical geometry, seen in Equation (4.7),

\[
\int_{r_w}^{r} k(T) dT = \frac{\dot{q} r_o^2}{4} \left[ 1 - \left( \frac{r}{r_o} \right)^2 \right].
\]  

Equation (4.7) cannot be solved explicitly for temperature until the temperature-dependent thermal conductivity is defined. Because of the complex nature of fuel behavior during irradiation, defining \( k(T) \) from in-pile instrumentation is also very difficult. Estimates of \( k(T) \) for irradiated nuclear fuel can be done in hot cells or by fuel modeling codes, but this approach may not account for important phenomena during irradiation. For example, the fuel pellet can swell and contact the cladding. This forms a rim around the pellet and can change material properties within the fuel rod, as seen in Figure 4-2.
Cracking, also seen in the cross-section image from Figure 4-2, is another example of changes that occur in fuel during irradiation that impact heat transfer through the fuel, since it is no longer a solid material. Another example is porosity gradients within the fuel, which is also seen in Figure 4-2. The porosity can vary from the pellet centerline to the outer rim causing changes in cross-sectional material properties.

4.1.3. Two-Thermocouple Testing Procedure and Measurement Sensitivity Testing

Using the definition from Equation (4.6), the required measured parameters to calculate thermal conductivity from the two-thermocouple method are: outer thermocouple distance from centerline, $r$; volumetric heat generation, $\dot{q}$; and measured temperature difference, $\Delta T$. The test setup shown in Figures 4-3 and 4-4 was used to obtain data for these parameters.
Figure 4-3. Theoretical test setup inside tube furnace.

Figure 4-4. Test setup at INL’s HTTL.
The samples were positioned inside a tube furnace to control ambient temperature and provide a sample temperature test range from 500 to 700 °C. A specified voltage and current was supplied to the sample by attaching connections from the power supply to each end of the rod using Inconel electrodes connected to Inconel clamps, as seen in Figure 4-5. Voltage measurement leads were attached to Inconel clamps at each end of the surrogate rod to measure the voltage drop in the sample. A precision current measurement measured current within the experimental test loop by applying Ohm’s law from measuring the voltage drop across the shunt and knowing the calibrated shunt resistance. Using the relationship between power, $P$, current, and voltage (e.g., $P = I \times V$), volumetric heat generation was calculated using the measured current, $I$, the measured voltage drop in the sample, $V$, and sample dimensions. Fluid flow within the tube was argon. Signals were processed by a data acquisition system to give temperature data from two type K thermocouples and power in the sample. The thermocouples were carefully positioned at known locations within the sample as seen in Figure 4-5.

Figure 4-5. CFOAM® sample with thermocouples inserted.
In this research, several test parameters were varied to estimate the sensitivity to measurement parameters of the steady-state method. Measurement sensitivity testing consisted of selecting variables to hold constant, such as power, and measuring values over the defined temperature range. For example, testing was conducted by holding supplied power at 100 watts and waiting for sample and furnace to reach steady-state equilibrium while maintaining constant argon flow rate within the tube. Once equilibrium is reached, the thermocouple readings do not change with respect to time, and data was recorded. Then, the ambient temperature was increased by increasing the tube furnace temperature to the desired setpoint temperature. The sample and furnace will again reach steady-state, and another measurement was taken. This process was repeated in order to generate data tables for each testing parameter. Table 4-1 shows measurement sensitivity parameters for steady-state method examinations.

4.1.4. Method Limitations and Uncertainty

There are acknowledged limitations to the two-thermocouple method. Placing two thermocouples within a prototypic-sized diameter fuel rod will incur significant perturbations in the measured fuel thermal conductivity and is not recommended for in-pile testing because of the geometry constraints listed in Section 2.1. However, the aim of this research is to provide understanding of method laboratory limitations only, and offer insights to further in-pile thermal conductivity advancements using variations of steady-state radial heat flow methods already used by the IFE/HRP.

4.1.4.1. Measurement Limitations

As mentioned in Section 4.1.5.1, electrical properties of the surrogate material deter-
mine the additional components of the setup. For example, if the electrical resistance of the material is low, more current is required to heat the sample. Large test currents require large power supplies and result in higher temperatures in connections and leads. Heat losses in connections and leads from resistance heating reduce the voltage potential across the sample; in turn, more current must be supplied to create the desired voltage drop across the sample. Materials with very low resistivity, are acceptable for use in components outside the furnace, but high temperature conditions limit the selection of materials in the furnace.

Electrical resistivity, which is a function of geometry through which the current passes, can help to prevent large heat losses by geometry selections. As seen in Equation (4.8), electrical resistance through a component can be changed by the length, area, or resistivity of the component.

\[
R = \frac{\rho_{\text{electrical}} L}{A},
\]

where:

\[
\begin{align*}
R & = \text{electrical resistance (Ohm)}, \\
\rho_{\text{electrical}} & = \text{static electrical resistivity (Ohm-m)}, \\
L & = \text{length of sample subjected to current (m), and} \\
A & = \text{area of sample subjected to current (m}^2) \]
\]

<table>
<thead>
<tr>
<th>Sensitivity</th>
<th>Experimental Parameter Varied</th>
</tr>
</thead>
<tbody>
<tr>
<td>Surrogate rod thermal conductivity</td>
<td>Surrogate material batch number</td>
</tr>
<tr>
<td>Ambient conditions for temperature-dependent thermal conductivity measurements</td>
<td>Furnace temperature (300-600 ºC)</td>
</tr>
<tr>
<td>Sample temperature gradient</td>
<td>Constant power setting</td>
</tr>
</tbody>
</table>
The application of Equation (4.8) helps to reduce the heat loss effects by effectively reducing resistance across components by geometry selections, but furnace space and muffle tube materials limit design geometry inside the furnace. Tube furnaces are convenient to regulate temperature, but are constrained to a specific test volume. Typically, the volume is enclosed in a 2 to 4 inch diameter muffle tube made of very high temperature materials, such as ceramics. Therefore, large diameter electrode rods or other heavy components cannot be employed in the furnace to support high current requirements.

Another limitation of the laboratory measurement is understanding the contact resistance between the thermocouple and the sample. Since the method uses two thermocouples, the error from this source is essentially doubled. As noted in Section 2.2.1 and by Tverberg [2] and Betten [16], great care is taken with in-pile thermal conductivity measurement methods to understand gap conductance in irradiated conditions between the thermocouple and fuel, as well as the fuel and the cladding. In the two-thermocouple evaluations, understanding or quantifying gap conductance is important since two thermocouples are used. For low temperature applications, a conductive grease or paste can be used to minimize the impact of gap resistance; however, most greases or pastes are limited to lower temperature limits, leaving few choices to enhance the conduction across the gap.

Initial efforts investigated the feasibility of inserting the thermocouple in the fuel before sintering as a method for reducing gap resistance between the sample and measurement sensor. Investigations were completed on cold-pressed zirconium diboride (ZrB₂) sintered at temperatures above 1300 °C. A hole was machined for the 1/16” thermocouple in the pre-sintered, “green” sample. As shown in Figure 4-6, ZrB₂ volumetric shrinkage after sintering was common in this research (as was previously observed in ZrB₂ research
efforts from Stucker [41] and Franke [42]). Post-sintering evaluations indicate that the thermocouples and sample were in intimate contact due to volumetric shrinkage that occurred during sintering at temperatures above 1300 ºC for 2 hours.

There are significant advantages of sintering the thermocouple in the fuel before irradiation experiments. Intimate contact greatly reduces uncertainties due to contact resistance and eliminates the need for drilling after the fuel is sintered. Where possible, it is recommended that sintering the thermocouple or hot wire probe into the fuel rod material be used in either the multiple thermocouple steady-state or transient methods.

4.1.4.2. Measurement Uncertainty Analysis

As a first effort, the common approach detailed by Beckwith et al. [43] was applied to estimate only the uncertainty in the experimental measurements (even though the approach and setup uncertainties are not included within this analysis of uncertainty). Equation (4.6) was rearranged to evaluate the uncertainty impact of each measurement parameter,

Figure 4-6. Sintered ZrB$_2$ sample in good contact with inserted thermocouple.
where \( \dot{q} \) is defined as the product of measured current, \( I \), and measured voltage, \( V \), divided by the volume, with \( r_o \) being the radius of the rod, and \( L \) the length of the rod. Defining \( dk \) as the uncertainty of Equation (4.6), the partial differentials can be taken of Equation (4.9)

\[
dk = \frac{V r^2}{4\pi\Delta T r_o^3 L} dI + \frac{I r^2}{4\pi\Delta T r_o^3 L} dV + \frac{2IVr}{4\pi\Delta T r_o^3 L} dr - \frac{IVr^2}{4\pi(\Delta T)^2 r_o^2 L} d\Delta T - \frac{IVr^2}{2\pi\Delta T r_o^3 L} dr_o - \frac{IVr^2}{4\pi\Delta T r_o^3 L^2} dL. \tag{4.10}
\]

Dividing by the definition of \( k \) given in Equation (4.9),

\[
\frac{dk}{k} = \frac{dV}{V} I + \frac{dI}{I} + 2\left(\frac{dr}{r} - \frac{dr_o}{r_o}\right) - \frac{d\Delta T}{\Delta T} - \frac{dL}{L}, \tag{4.11}
\]

Using uncertainty terminology from Beckwith et al., where

\[
\varepsilon_k = \frac{dk}{k}, \varepsilon_V = \frac{dV}{V}, \varepsilon_I = \frac{dI}{I}, \ldots, \varepsilon_L = \frac{dL}{L}. \tag{4.12}
\]

Thus, a first approximation for the general measurement uncertainty of the two-thermocouple experimental method is

\[
\varepsilon_k = \sqrt{(\varepsilon_V)^2 + (\varepsilon_I)^2 + 2(\varepsilon_r)^2 + 2(\varepsilon_{r_o})^2 + (\varepsilon_{\Delta T})^2 + (\varepsilon_L)^2}. \tag{4.13}
\]

In the above equation, \( \varepsilon_V \) is the voltage measurement uncertainty from the power supply manufacturer, \( \varepsilon_I \) is the current measurement uncertainty based on calibration numbers, \( \varepsilon_r \) is the uncertainty from radial distance measurement, \( \varepsilon_{\Delta T} \) is the uncertainty from the \( \Delta T \) measurement given by the thermocouple manufacturer, \( \varepsilon_{r_o} \) is the radius measurement uncertainty, and \( \varepsilon_L \) is the length measurement uncertainty. Table 4-2 shows the percentage of contributing uncertainty from each of these sources, and indicates that the largest calcu-
lated uncertainty is from placement of the thermocouples within the sample and measuring the exact location. The assumption of measuring temperature at a finite point within the material of the rod is used with this method. This assumption does not include the thermocouples having a different material than the surrogate rod material; therefore, more error will be introduced with increasing thermocouple diameter. Also, there can be a considerable error contribution by gap resistance between the thermocouple and the sample.

### 4.1.5. Two-Thermocouple Method Surrogate Material

Investigations used a surrogate material in a laboratory setting. There are several advantages to this approach. A surrogate material is an effective and inexpensive tool to gain understanding about the viability of the two-thermocouple method. Thorough investigations can be completed to understand the method’s limitations and capabilities. In addition, results can be extrapolated to gain insights about potential in-pile applications.

Spreadsheets were created with upper and lower measurement limits of current, volt-

<table>
<thead>
<tr>
<th>Error Source</th>
<th>Error Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\varepsilon_V$</td>
<td>2.10</td>
</tr>
<tr>
<td>$\varepsilon_I$</td>
<td>0.10</td>
</tr>
<tr>
<td>$\varepsilon_r$</td>
<td>8.36</td>
</tr>
<tr>
<td>$\varepsilon_{\Delta T}$</td>
<td>0.75</td>
</tr>
<tr>
<td>$\varepsilon_{r_o}$</td>
<td>0.10</td>
</tr>
<tr>
<td>$\varepsilon_L$</td>
<td>9.30E-2</td>
</tr>
<tr>
<td>$\varepsilon_k$</td>
<td>12.03</td>
</tr>
</tbody>
</table>
age, electrical resistance, thermal conductivity, sample geometry, and sample temperature difference. From these spreadsheets, a limiting range was calculated for selecting possible materials. Machinability and maximum service temperature were some of the other important selection criteria.

4.1.5.1. Desired Two-Thermocouple Method Surrogate Characteristics

Because the goal of laboratory tests is to investigate methods for in-pile use, the surrogate material chosen should exhibit certain characteristics.

- Thermal conductivity close to that of UO\textsubscript{2} fuel, ranging around 2 to 10 W/m°C over a temperature range of 100 to 1600 °C

Selecting a surrogate material with thermal conductivity in the range of 3 to 8 W/m°C, determines the method’s ability to measure thermal conductivity in the range of the fuel.

- Detect thermal conductivity changes with temperature

The surrogate material thermal conductivity change as a function of temperature is beneficial. The thermal conductivity of UO\textsubscript{2} degrades with temperature and rod burnup, (e.g., see References [2], [4], and [16]). To validate the technique, it is important measure differences in thermal conductivity readings as testing parameters are changed.

- Electrically resistive material to support Joule Heating

As mentioned, the two-thermocouple method uses Joule Heating to simulate volumetric heat generation; thus, current must flow through the sample. The resistance of the material determines how well the heat will build up in the sample. Surrogate materials with large resistance values are desired in the two-thermocouple method.
• Material able to support large temperature drop (\( \Delta T > 5^\circ C \))

It is important that the measured temperature difference, \( \Delta T \), is larger than the potential thermocouple uncertainty. It is ideal for \( \Delta T \) to be as large as possible to understand the benefits and limitations of the two-thermocouple method; however, the temperature drop in the sample is dependent on constant supplied power.

• Material for high temperature use

Laboratory testing is completed in a high temperature controlled furnace while controlling the ambient environment inside the furnace with air or an inert gas. Selecting materials to withstand the demands of temperature in air and inert conditions is desired.

• Material easily machined and durable

Besides machining rod geometries and thermocouple holes, specific high-tolerance material property samples must be machined for temperature-dependent thermophysical measurements. Also, samples are periodically removed from testing for inspection and adjustments, and subjected to repeated high temperatures for long durations. These requirements call for a surrogate material to withstand temperature cycles, be machinable, and withstand experimentalist handling.

4.1.5.2. CFOAM®

The surrogate fuel rod material chosen for research effort is CFOAM®, manufactured by Touchtone Research Laboratories Ltd. [44]. This carbon structural foam is non-combustible and will not off-gas at high temperatures. CFOAM® is calcined coke (CAS #64743-05-1) engineered to meet high performance material needs. Table 4-3 summarizes properties provided by Touchtone Research Laboratories Ltd for two types of CFOAM®,
Although both CFOAM® materials (CFOAM20 and CFOAM25) were initially considered, the denser CFOAM25 had more desirable properties for this application. Two CFOAM25 samples from different batches were tested for measurement repeatability.

### 4.1.5.3. Surrogate Material Properties Measurements

Initial room temperature material property data for CFOAM25 suggested that it would be suitable for testing in a laboratory setting. However, in order to properly validate the steady-state method for estimating thermal conductivity, temperature-dependent properties of CFOAM25 must be quantified because its thermal conductivity is not well defined. Three important property measurements were made to estimate the material's temperature-
dependent thermal conductivity. Thermal conductivity of a material can be defined as the product of density, specific heat, and thermal diffusivity. These values were measured using specialized laboratory systems located at INL’s HTTL.

**Density**

The density of the surrogate material as a function of temperature was calculated using data obtained from a pushrod dilatometer. This machine measures thermal elongation of a material with respect to temperature. Recalling that density is fundamentally defined as mass per volume, the linear coefficient of thermal expansion is defined as the differential change in length per change in temperature,

\[ \alpha_L = \frac{1}{L_0} \frac{\partial L}{\partial T}, \]  

(4.14)

where \( L_0 \) is the initial length. The above expression is often rewritten as

\[ \frac{\Delta L}{L} = \alpha_L \Delta T, \]  

(4.15)

where, \( \Delta L \) is the sample change in length, \( \alpha_L \) is the coefficient of linear expansion, and \( \Delta T \) is the sample change in temperature. For isotropic materials, the volumetric coefficient of expansion is very closely approximated as three times the linear coefficient of thermal expansion,

\[ \frac{\Delta V}{V_0} = 3 \alpha_L \Delta T, \]  

(4.16)

where \( \Delta V \) is the sample volume change, and \( V_0 \) is the initial volume. The final volume is defined as initial volume plus the change in volume. These relationships can be combined to obtain the final density, defined as a function of sample mass, initial sample volume and
length, and change in sample length, as shown by Equation 4.17,

\[
\rho_f = \frac{\text{mass}}{V_\alpha \left(1 + 3 \frac{\Delta L}{L_0}\right)}.
\]

(4.17)

**Thermal diffusivity**

Thermal diffusivity, \( \alpha \), is defined as the material’s thermal conductivity divided by the product of the material’s density and specific heat. Hence, thermal diffusivity effectively relates a material’s ability to conduct energy to its ability to store energy. The thermal diffusivity of CFOAM25 was measured at INL’s HTTL using a laser flash thermal diffusivity system. The system provides high energy pulse heating to one surface of a sample; the imposed thermal transient allows measurements of how well heat transfers through the sample, which is then used to estimate the material’s thermal diffusivity.

**Specific heat capacity**

Specific heat capacity measurements were conducted at INL’s HTTL using a Differential Scanning Calorimeter (DSC). A complete DSC test requires three individual tests: a baseline test void of any test material (results from this test are used to eliminate any bias from test to test variations), a test containing a reference sample with well known \( C_p \) values in order to calculate the unknown sample \( C_p \) values, and a test with sample whose properties are unknown. Precision is required for accurately characterizing the specific heat using this test, and one of the more important requirements is closely matching the masses of the test sample to the reference sample.

**Thermal conductivity**

Once the above measurements are made for the surrogate material, its temperature-dependent thermal conductivity from material properties measurements was estimated
using Equation 4.18,

\[ k = \alpha \rho C_p. \] (4.18)

4.2. Steady-State Two-Thermocouple Method Summary

Steady-state testing followed the procedure outlined in Section 4.1.3 to generate thermal conductivity measurement results for the surrogate material over the test temperature range of 500 to 700 ºC. These results (details provided in Chapter 6) were compared to the surrogate material thermal conductivity temperature-dependent estimates from specific laboratory properties measurements (e.g., density, specific heat capacitance, and thermal diffusivity described in Section 4.1.5.3). Measurement sensitivity parameters detailed in Section 4.1.3 were investigated to determine the accuracy and limitations of the steady-state method, with the results from these sensitivities tests also provided in Chapter 6.
CHAPTER 5
TRANSIENT METHOD APPROACH AND PROCEDURE

Chapter 5 provides details about the setup and procedure for the transient method. The section discusses relevant background information with governing equations, measurement uncertainties, and surrogate materials used for method evaluation.

5.1. Needle Probe Transient Method Background and Governing Equations

Carslaw and Jaeger [20] and Wechsler [45] state that the temperature rise at any point in a material resulting from an internal heat source is dependent on the material thermal conductivity. 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 equilibrium, 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 5-1 (linear region of the time period 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 in an infinite solid. The analytical representation, from the derivation given in Section 2.3.1, of this relationship for a long duration heating time solution is given by Carslaw and Jaeger [20], is given in Equation (5.1) for $0 < t < t_{\text{heating}}$
\[ T_{r2} - T_{r1} = -\frac{Q'}{4\pi k} Ei\left(\frac{r^2}{4\alpha t}\right), \]  

(5.1)

where:
- \( t \) = time (s),
- \( T_{r1} \) = probe center temperature (°C),
- \( T_{r2} \) = temperature at probe radius (°C),
- \( Q' \) = heat input per unit length (W/m),
- \( r \) = distance from the heat source (m),
- \( \alpha \) = thermal diffusivity of the sample (m²/s),
- \( k \) = thermal conductivity of the sample (W/m°C),
- \( t_{\text{heating}} \) = total heating time (s), and
- \( Ei \) = first order exponential integral function\(^1\) defined as:

\[ Ei(x) = \int_{x}^{\infty} e^{-t} dt, \]  

(5.2)

or in series form for \( x > 0 \):

\(^1\) The exponential integral is a commonly used non-elementary mathematical function, see Kreyszig [46].
where, $\gamma$, is the Euler–Mascheroni constant approximated to four decimal places as 0.5772. Thus, Equation (5.1) becomes

$$T_{r_2} - T_{r_1} = -\frac{Q'}{4\pi k} \left[ \gamma + \ln\left(\frac{r_2^2}{4\alpha t}\right) - \frac{r_2^2}{4\alpha t} + \frac{r_4^4}{64\alpha^2 t^2} + \ldots \right].$$  \quad (5.4)

Equation (5.4) is dependent on the material properties, geometry of the sample to be measured, and the test conditions (e.g., heat input and heating time). For large values of time, the higher order terms in Equation (5.4) are negligible. Thus, for a fixed probe radius, $r$, measurement times, $t_1$ and $t_2$, and temperatures, $T_1$ and $T_2$, identified from the linear region in Figure 5-1, Equation (5.4) is simplified to the Carslaw and Jaeger derived long time solution shown in Equation (5.5)

$$T_2 - T_1 = \frac{Q'}{4\pi k} \ln\left(\frac{t_2}{t_1}\right).$$  \quad (5.5)

It is shown in Figure 5-1, that the temperature increase is linearly related to the temperature versus the natural logarithm of time by the mathematical relationship to define the slope, $S$, as given by Equation (5.6)

$$S = \frac{(T_2 - T_1)}{\ln\left(\frac{t_2}{t_1}\right)}. \quad (5.6)$$

Equation (5.6) is combined with Equation (5.5) providing the relationship shown in Equation (5.7) to calculate the thermal conductivity that is given in ASTM Standard D 5334-08

$$k = \frac{CQ'}{4\pi S}, \quad (5.7)$$
where:

\[ C = \text{calibration constant}, \]
\[ Q' = \text{heat input by the line heater per unit length (W/m), and} \]
\[ S = \text{linear slope of the temperature versus the natural logarithm of time (°C), defined by Equation (5.6).} \]

The calibration constant suggested by ASTM Standard D 5334-08 in Equation (5.7), \( C \), is simply the ratio of a known reference material thermal conductivity to that of the measured thermal conductivity calculated from the needle probe method. This constant accounts for the finite size of the heater and differences in properties between the sample, line heater, and the thermocouple. The slope, \( S \), of the identified linear region of the temperature versus the natural logarithm of time is calculated by Equation (5.6). The heat input per unit length of the line heater, \( Q' \), is related to the thermocouple temperature at the time when the temperature versus the natural logarithm of time response curve starts to become linear (\( T_1 \) as seen in Figure 5-1) and the temperature when the response curve ceases to be linear (\( T_2 \) in Figure 5-1). The equation to calculate \( Q \) is given by

\[ Q' = \frac{VI}{L}, \quad (5.8) \]

where:

\[ V = \text{measured voltage drop of the line heater contained in the probe (V)}, \]
\[ I = \text{measured current (amps), and} \]
\[ L = \text{length of the heater wire (m)}. \]

A similar relationship can be applied to calculate the thermal conductivity of a cooling sample with the heat source removed.

5.2. Needle Probe Design and Component Discussion

General details of the thermal probe design are provided in this section, as well as specific component discussions with respect to candidate materials for in-pile application.
Careful consideration of material property requirements must be made. This includes consideration of thermal and electrical properties, irradiation resistance, and material compatibility requirements. Section 5.2 contains probe design requirements for each component of the needle probe, including the thermocouple, insulation, heater wire, and sheath.

A schematic of a general probe design for room temperature application is given in Figure 5-2. As suggested by ASTM D 5334-08 [31], a wire heating element loops within a sheathed probe. The heating wire is continuous and connected at the end of the probe to a power source to supply either constant current or constant power. A thermocouple is also within the sheathed probe. The thermocouple junction is positioned at half the heater section length and the thermocouple leads extend from the probe to a data acquisition system. Figure 5-3 shows a cross section view (location A-A of Figure 5-2) of only the heater section of a probe after swaging that only contains the thermoelements. Figure 5-4 shows a cross section at location B-B that contains the heater wires and the thermocouple wires. Maintaining spacing between the heater wires and thermocouple wires during fabrication is important for continuity and probe longevity.

Figure 5-2. Room temperature thermal probe schematic with ceramic insulation fill.
Figure 5-3. Cross-sectional view A-A of swaged probe containing heater wires only.

Figure 5-4. Cross-sectional view B-B of swaged probe containing heater wires and thermocouple wires.
5.2.1. Thermocouple

For many applications, a type K thermocouple may be used. However, for high temperature irradiation conditions, an INL-developed doped molybdenum/niobium alloy High Temperature Irradiation Resistant ThermoCouple (HTIR-TC) is recommended. This thermocouple allows precise measurements in high temperature, irradiation conditions without decalibration due to transmutation [47]. However, specific in-pile test conditions will ultimately dictate the thermocouple selection.

5.2.2. Insulation

Design requirements for the probe insulation include:

- separation of wires contained in the probe (both legs of the heater wire and the thermocouple wires),
- electrical insulation,
- effective heat transfer from the heater to the sample,
- high temperature applications,
- irradiation resistant, and
- materials compatibility between probe sheath and wires at high temperature.

Ceramic insulators were selected because of their thermal and electrical properties. Candidate insulators considered include: alumina (Al₂O₃), beryllia (BeO)¹, hafnia (HfO₂), zirconia (ZrO₂), and magnesia (MgO). All materials have very good electrical insulation properties and can be used in reactor needle probe because of their capacity to insulate

¹ BeO is toxic in powder form and can cause Berylliosis, a chronic lung disease. Therefore, crushable BeO insulators are recommended with extreme caution. In addition, crushable BeO insulators are expensive and difficult to find because of limited available vendors.
electrical signals over a wide temperature range. Electrical insulation values are greatly affected by material impurities, and certified high purity (e.g., > 99.9%) ceramic insulators are recommended [48]. The electrical insulation capability of ceramic insulators degrades when subjected to high temperatures, but values are generally large enough to provide electrical insulation benefits for most in-pile applications and temperatures, as seen in Figure 5-5 [49].

High insulator thermal conductivity is beneficial to efficiently transfer heat to the sample as it is dissipated by the heater wires. Low ceramic insulator thermal conductivity will cause heat build up within the probe, and delay probe response time to the sample. Candidate ceramic insulator thermal conductivities as a function of temperature are given in Figure 5-6 [50].

![Figure 5-5. Electrical resistivity of selected ceramic insulation materials as a function of temperature [49].](image)

**Figure 5-5.** Electrical resistivity of selected ceramic insulation materials as a function of temperature [49].
Another important consideration is the effect of densification on ceramic insulator thermal conductivity. The thermal conductivity of ceramic insulators can vary dramatically with theoretical density, where in some cases even a 5% change in theoretical density can vary thermal conductivity by 70-80% of the reported value for a fully dense material [48]. The most effective method to control density and probe diameter is done by swaging. Swaging also minimizes the contact resistance in the probe between the insulation and the heater wires, insulation and the thermocouple wires, and insulation and the sheath.

Insulator materials for in-pile needle probe application must be selected primarily on test conditions and material availability. Because testing conditions will vary depending on specific measurement needs, insulation material selections should be based on maxi-
mum temperature testing conditions, materials used in the probe, and required thermal and electrical properties.

5.2.3. Heater Wire

The heater wire is an important component of the design and careful selection is critical. Some of the important considerations are melting temperature, maximum recommended working temperature, thermal neutron capture cross section, electrical resistivity, material workability and compatibility with candidate insulation.

5.2.3.1. Temperature Limitations

The target maximum probe temperature for experimental in-pile testing is 1800 °C, thus melting temperatures and maximum recommended working temperatures must be considered in material selection. Some candidate high temperature wire melting temperature and maximum recommended working temperatures are given in Table 5-1.

Table 5-1. High Temperature Wire Properties [48]

<table>
<thead>
<tr>
<th>Material</th>
<th>Melting Temperature (°C)</th>
<th>Maximum Recommended Working Temperature (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Molybdenum</td>
<td>2610</td>
<td>1900</td>
</tr>
<tr>
<td>Tungsten</td>
<td>3380</td>
<td>2200</td>
</tr>
<tr>
<td>Niobium</td>
<td>2468</td>
<td>1800</td>
</tr>
<tr>
<td>Tantalum</td>
<td>2996</td>
<td>2400</td>
</tr>
<tr>
<td>Rhenium</td>
<td>3180</td>
<td>2400</td>
</tr>
<tr>
<td>Chromium</td>
<td>1907</td>
<td>1400&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td>Nickel</td>
<td>1453</td>
<td>1100&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
</tbody>
</table>

<sup>a</sup>: Estimated
All heater materials listed in Table 5-1 meet the target maximum testing temperature and are recommended for very high temperature heater wire materials, except for nickel and chromium because of the recommended maximum working temperature value. However, heater wire material selections are dependent on applicational need for specific testing conditions.

5.2.3.2. *Thermal Neutron Capture Cross Section*

Transmutation of elements under irradiation can be detrimental to experiment design as assumed material temperature-dependent properties and thermocouple calibration curves are no longer applicable. Material selection can help minimize transmutation. Table 5-2 compares the thermal neutron capture cross-section of candidate needle probe materials.

5.2.3.3. *Electrical Resistivity*

As seen in Equation (4.8), the resistance of a material is a function of resistivity. The needle probe method at high temperatures relies on knowing the resistance of the heater wire, which has temperature dependence on resistivity. Therefore, quantifying the resistivity change as a function of temperature will greatly increase probe accuracy. More resistance heating will occur from a higher resistivity value. Selecting a material with relatively high electrical resistivity at high temperatures for the heater wire is advantageous to the design. Electrical resistivities as a function of temperature of selected candidate heater wire materials are shown in Figure 5-7.
Table 5-2. Thermal Neutron Capture Cross Section of Candidate Wire Materials [48]

<table>
<thead>
<tr>
<th>Element</th>
<th>Thermal neutron capture cross section for 2200 m/sec. (barns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Beryllium</td>
<td>0.01</td>
</tr>
<tr>
<td>Magnesium</td>
<td>0.06</td>
</tr>
<tr>
<td>Zirconium</td>
<td>0.18</td>
</tr>
<tr>
<td>Aluminum</td>
<td>0.23</td>
</tr>
<tr>
<td>Niobium</td>
<td>1.1</td>
</tr>
<tr>
<td>Molybdenum</td>
<td>2.5</td>
</tr>
<tr>
<td>Chromium</td>
<td>2.9</td>
</tr>
<tr>
<td>Nickel</td>
<td>4.6</td>
</tr>
<tr>
<td>Tungsten</td>
<td>19.2</td>
</tr>
<tr>
<td>Tantalum</td>
<td>21.3</td>
</tr>
<tr>
<td>Hafnium</td>
<td>105</td>
</tr>
</tbody>
</table>

Figure 5-7. Temperature-dependent electrical resistivities of Inconel [51], tantalum, niobium, nickel, tungsten, and molybdenum [52].
5.2.3.4. Material Workability and Compatibility

Material workability and compatibility require probe design consideration for in-pile applications. For example, the probe design calls for a heater element wire to make a 180º bend in the bottom of the probe. This 180º bend eliminates brittle materials from very small probe diameters. Also, the heater wire material must exhibit ductility when subjected to swaging passes to reduce outside diameter. Table 5-3 gives maximum compatibility temperatures for candidate metals and insulation materials.

Table 5-3. Maximum Compatibility Temperature for Various Materials

<table>
<thead>
<tr>
<th>Material</th>
<th>Mo</th>
<th>Nb</th>
<th>Pt</th>
<th>Re</th>
<th>Rh</th>
<th>Ta</th>
<th>Ti</th>
<th>W</th>
<th>Zr</th>
</tr>
</thead>
<tbody>
<tr>
<td>Insulators</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Alumina</td>
<td>&gt;1800</td>
<td>&gt;1800</td>
<td>&gt;1770</td>
<td>&gt;2000</td>
<td>&gt;1960</td>
<td>&gt;1650</td>
<td>&gt;1650</td>
<td>&gt;1900</td>
<td>&gt;1200</td>
</tr>
<tr>
<td>Hafnia</td>
<td>&gt;2200</td>
<td>&gt;2200</td>
<td>&gt;1770</td>
<td>&gt;2200</td>
<td>&gt;1960</td>
<td>&gt;1872</td>
<td>&gt;1650</td>
<td>&gt;1700</td>
<td>&gt;1800</td>
</tr>
<tr>
<td>Beryllia</td>
<td>&gt;1900</td>
<td>&gt;1800</td>
<td>&gt;2570</td>
<td>&gt;2400</td>
<td>&gt;2100</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Zirconia</td>
<td>&gt;1900</td>
<td>&gt;1600</td>
<td>&gt;1770</td>
<td>&gt;1700</td>
<td>&gt;1960</td>
<td>&gt;1790</td>
<td>&gt;1650</td>
<td>&gt;1700</td>
<td>&gt;1800</td>
</tr>
<tr>
<td>Metals</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Molybdenum</td>
<td>NA</td>
<td>&gt;2470</td>
<td>&gt;1770</td>
<td>&gt;2510</td>
<td>&gt;1940</td>
<td>&gt;2610</td>
<td>&gt;1670</td>
<td>&gt;2470</td>
<td>&gt;1550</td>
</tr>
<tr>
<td>Niobium</td>
<td>&gt;2470</td>
<td>NA</td>
<td>&gt;1700</td>
<td>&gt;2160</td>
<td>&gt;1500</td>
<td>&gt;2470</td>
<td>&gt;1670</td>
<td>&gt;2470</td>
<td>&gt;1740</td>
</tr>
<tr>
<td>Platinum</td>
<td>&gt;1770</td>
<td>&gt;1700</td>
<td>NA</td>
<td>&gt;1770</td>
<td>&gt;1770</td>
<td>&gt;1760</td>
<td>&gt;1310</td>
<td>&gt;1770</td>
<td>&gt;1150</td>
</tr>
<tr>
<td>Rhodium</td>
<td>&gt;1940</td>
<td>&gt;1500</td>
<td>&gt;1770</td>
<td>&gt;1960</td>
<td>NA</td>
<td>&gt;1740</td>
<td>&gt;1300</td>
<td>&gt;1960</td>
<td>&gt;1070</td>
</tr>
<tr>
<td>Tantalum</td>
<td>&gt;2610</td>
<td>&gt;2470</td>
<td>&gt;1760</td>
<td>&gt;2690</td>
<td>&gt;1740</td>
<td>NA</td>
<td>&gt;1668</td>
<td>&gt;3017</td>
<td>&gt;1852</td>
</tr>
<tr>
<td>Titanium</td>
<td>&gt;1670</td>
<td>&gt;1670</td>
<td>&gt;1310</td>
<td>&gt;1670</td>
<td>&gt;1300</td>
<td>&gt;1668</td>
<td>NA</td>
<td>&gt;1670</td>
<td>&gt;1540</td>
</tr>
<tr>
<td>Tungsten</td>
<td>&gt;2470</td>
<td>&gt;2470</td>
<td>&gt;1770</td>
<td>&gt;2825</td>
<td>&gt;1960</td>
<td>&gt;3017</td>
<td>&gt;1670</td>
<td>NA</td>
<td>&gt;1735</td>
</tr>
<tr>
<td>Zirconium</td>
<td>&gt;1550</td>
<td>&gt;1740</td>
<td>&gt;1150</td>
<td>&gt;1590</td>
<td>&gt;1070</td>
<td>&gt;1852</td>
<td>&gt;1540</td>
<td>&gt;1735</td>
<td>NA</td>
</tr>
</tbody>
</table>

a. See References [10] and [48]
b. See Reference [48]
c. Not applicable
Compatibility of heater wire and insulation materials at high temperatures is another important consideration as in-pile reactions may alter chemical compositions, change electric output, or experience eutectic interactions.

The probe design requires workable materials for effective probe fabrication because of small geometry requirements. Thus, care must be taken to select heater wire materials which are compatible and workable. Table 5-4 from Rempe et al. [10], provides information about oxidation resistance and general metals workability notes, such as, machinability, weldability, and additional material comments.

Wire materials such as, niobium and molybdenum offer the best wide range capabilities, and several material combinations are acceptable for in-pile application; however, specific testing conditions will ultimately dictate heater wire material selections.

Table 5-4. Heater Wire Material Workability Properties and Notes

<table>
<thead>
<tr>
<th>Material</th>
<th>Oxidation Resistance&lt;sup&gt;a&lt;/sup&gt;</th>
<th>Material Workability Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Molybdenum</td>
<td>No</td>
<td>Welded conventionally, difficult machining</td>
</tr>
<tr>
<td>Niobium</td>
<td>No</td>
<td>Ductile, weldable, machinable</td>
</tr>
<tr>
<td>Platinum</td>
<td>Yes</td>
<td>Ductile, rare material, expensive</td>
</tr>
<tr>
<td>Rhenium</td>
<td>No</td>
<td>Ductile, weldable, not easily machined, used mainly in alloy form, expensive</td>
</tr>
<tr>
<td>Rhodium</td>
<td>Yes</td>
<td>Expensive</td>
</tr>
<tr>
<td>Tantalum</td>
<td>No</td>
<td>Ductile, weldable</td>
</tr>
<tr>
<td>Titanium</td>
<td>No</td>
<td>Ductile, weldable, machinable, widely used</td>
</tr>
<tr>
<td>Tungsten</td>
<td>No</td>
<td>Not easily machined or welded</td>
</tr>
<tr>
<td>Zirconium</td>
<td>No</td>
<td>Machinable and weldable with cover gas</td>
</tr>
</tbody>
</table>

<sup>a</sup> See Reference [10] for oxidation information
5.2.4. Sheath Material

The needle probe sheath materials must exhibit certain characteristics and material properties for in-pile use, such as:

- low neutron absorption values (see selected elements listed in Table 5-2),
- compatibility between ceramic insulation with respect to high temperature interactions and thermal expansion coefficients,
- workable, weldable, and ductile for swaging and leak tight fabrication,
- and high thermal conductivity to effectively transfer heat to the sample.

Stainless steel, Inconel, and niobium 1% Zr sheaths are commonly used materials for reactor thermocouple applications and are recommended for probe designs.

5.2.5. Probe Component Design Conclusions

Recommendations found in Sections 5.2.1 and 5.2.4 are helpful to select appropriate materials for probe components. As mentioned in these sections, ultimately, specific in-pile testing conditions will direct material selections to optimize probe components. This research has investigated probe designs for laboratory testing at room temperature to 400 °C with the intent to optimize probe component material properties capable of in-pile applications. For example, for room temperature evaluations, a type K thermocouple and lower temperature heater wires, insulation, and sheath materials were be used. Higher temperature design required higher temperature material selections for probe components. An invention disclosure, filed by USU and INL details materials, geometry, and fabrication details used for these probe designs.

Two probes were designed and fabricated for thermal conductivity measurements. A
room temperature probe for initial concept evaluation and a high temperature probe for specific elevated temperature testing. For labeling purposes, the room temperature probe is referenced as the “RT probe”, and the high temperature probe is referenced as the “HT probe” in this thesis.

5.3. Needle Probe Testing Procedure

The required measurement parameters from Equation (5.7) are supplied power to the heater, sample geometry, slope of the time-temperature plot (as illustrated in Figure 5-1), and the calibration constant (if needed). The power supplied to the probe/sample is known by measuring the voltage drop in the probe with voltage measurement leads at the probe’s end and calculating the constant current in the loop by measuring the voltage drop across a precision resistor, commonly called a current shunt, in one of the power supply leads. From the relationship of \( P = I \times V \), power is calculated. A LabVIEW® program to regulate constant power was used. The program adjusted the output voltage to maintain the set-point power setting. A switch in the program controlled the current flow from the power supply to the probe, and this was used to quickly apply or turn off power to the sample. Leads to monitor the temperature response of the probe were attached to the thermocouple wires at the probe’s end. All measurement leads were connected to the data acquisition system for data collection.

The needle probe method was tested at room temperature and elevated temperatures to view the accuracy and limitations of the method.

5.3.1. Room Temperature Needle Probe Test Setup

The room temperature setup seen in Figure 5-8, shows the probe in the sample. The
needle probe setup for room temperature experiments were used to assess the viability of further temperature-dependent testing. Thus, all materials and connections for room temperature testing are temperature limited. Connections to the power supply are made using the upper set of red and black clips. The other set of red and black clips are to measure the sample voltage drop close to the probe exit. This measurement is essential for quantifying the heat applied in the probe. The type K (yellow) connector shown in this figure is for the thermocouple leads.

5.3.2. Temperature-Dependent Needle Probe Setup

The temperature-dependent setup, seen in the schematic in Figure 5-9, is similar to the room temperature setup except a tube furnace is used to control ambient temperature.
Connections to thermocouple leads and heater wire to supply current and measure probe voltage drop are similar to those seen in Figure 5-8. The tube furnace is controlled at a desired temperature and monitored by a National Institute of Standards and Testing (NIST) traceable Type S thermocouple. The measured probe voltage drop is divided into several sections to account for the temperature-dependent electrical resistivity of the heater wire. Resistance in the heating section can be calculated; and from this, the power supplied to the sample can be estimated. The temperature-dependent setup at INL’s HTTL is given in Figure 5-10.

5.3.3. Tube Furnace Temperature Profile

When the probe is inserted into the furnace (as shown in Figure 5-10), it is subjected to a temperature gradient that varies from the furnace setpoint temperature to room temperature (where the probe connections are located). At the desired setpoint temperature, a
NIST-traceable Type S thermocouple was positioned at the furnace center while the furnace was allowed to reach a steady-state. At one inch increments from the furnace center to the furnace exit, the temperature distribution in the furnace was profiled to accurately estimate the temperatures experienced by the heater leads (and hence, their temperature-dependent resistance). An alumina muffle tube extends 6 inches from the furnace exit. The distance from the furnace center to furnace exit was 20 inches. The tube furnace profiles for setpoint temperatures of 250 and 400 °C are shown in Figure 5-11, as well as an assumed linear profile from the furnace exit to room temperature.
5.4. Needle Probe Method Testing Parameters and Measurement Uncertainty

In these evaluations, several test parameters are varied to estimate the accuracy of the needle probe method. Table 5-5 shows the proposed test matrix.

Table 5-5. Transient Needle Probe Method Measurement Sensitivity Parameters

<table>
<thead>
<tr>
<th>Sensitivity</th>
<th>Experimental Parameter Varied</th>
</tr>
</thead>
<tbody>
<tr>
<td>Material conductivity</td>
<td>Surrogate material</td>
</tr>
<tr>
<td>Power supply response</td>
<td>Power supply parameters</td>
</tr>
<tr>
<td>Temperature dependence</td>
<td>Tube furnace setpoint temperature (25 - 400°C)</td>
</tr>
<tr>
<td>Probe to surrogate rod contact</td>
<td>Add graphite powders or thermal grease</td>
</tr>
</tbody>
</table>

Figure 5-11. Furnace profile for 250 ºC and 400 ºC.
As a first effort, the approach Beckwith et al. [43] was applied to estimate the uncertainty in the experimental measurements. Equations (5.7) through (5.8) are rearranged to evaluate the uncertainty impact of each measurement parameter using a similar approach detailed in Section 4.1.4.2. Table 5-6 shows the percentage from each measurement uncertainty source, where $\varepsilon_V$ is the voltage measurement uncertainty from the power supply manufacturer, $\varepsilon_I$ is the current measurement uncertainty based on calibration numbers, $\varepsilon_S$ is the slope calculation uncertainty, and $\varepsilon_L$ is the length measurement uncertainty.

A value of $\varepsilon_S$ was chosen to be 5% for this analysis; however, with the use of linear regression techniques, the contribution of error from $\varepsilon_S$ can be much lower than the value of 5% selected. With a conservative value selected for $\varepsilon_S$, the total measurement uncertainty for $\varepsilon_k$ is well within acceptable limits for experimental analysis.

### 5.5. Needle Probe Surrogate Materials

Investigations use a surrogate material in a laboratory setting to investigate the needle probe method accuracy and limitations. These surrogate materials allow a low cost assessment method accuracies and probe design viability.

Table 5-6. Needle Probe Method Measurement Contributing Error

<table>
<thead>
<tr>
<th>Error Source</th>
<th>Error Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\varepsilon_V$</td>
<td>2.10</td>
</tr>
<tr>
<td>$\varepsilon_I$</td>
<td>0.10</td>
</tr>
<tr>
<td>$\varepsilon_S$</td>
<td>5</td>
</tr>
<tr>
<td>$\varepsilon_L$</td>
<td>9.30E-2</td>
</tr>
<tr>
<td>$\varepsilon_k$</td>
<td>5.43</td>
</tr>
</tbody>
</table>
5.5.1. Room Temperature Surrogate Materials

Surrogate materials for the needle probe method were first selected for room temperature method evaluations. Table 5-7 lists selected surrogate materials with their reported room temperature thermal conductivity values. An average, as well as the standard deviation from the average of all reported values, can also be found in Table 5-7. Fused Silica was the selected ASTM reference material. The five room temperature surrogate materials chosen for needle probe method testing had diameters between 1 and 2 inches and lengths between 6.5 and 7 inches. It must be noted to the reader that exact thermal conductivity

<table>
<thead>
<tr>
<th>Material</th>
<th>Reported Minimum Thermal Conductivity (W/m·ºC)</th>
<th>Reported Maximum Thermal Conductivity (W/m·ºC)</th>
<th>Average Reported Thermal Conductivity (W/m·ºC)</th>
<th>Reported Standard Deviation (W/m·ºC)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Delrin Plastic a</td>
<td>0.23</td>
<td>0.38</td>
<td>0.34</td>
<td>0.05</td>
</tr>
<tr>
<td>Acrylic b</td>
<td>0.17</td>
<td>0.25</td>
<td>0.20</td>
<td>0.02</td>
</tr>
<tr>
<td>Particle Board c</td>
<td>0.08</td>
<td>0.17</td>
<td>0.13</td>
<td>0.03</td>
</tr>
<tr>
<td>Polycarbonate d</td>
<td>0.19</td>
<td>0.22</td>
<td>0.20</td>
<td>0.01</td>
</tr>
<tr>
<td>SiO₂</td>
<td>Touloukian Recommended Value: 1.37 [50]</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

a. For reported low value, see: http://www.engineersedge.com/plastic/materials_common_plastic.htm; for reported high value, see: http://plastics.dupont.com/plastics/pdf/tit/americas/delrin/230323c.pdf

b. For reported low and high values, see: http://www.efunda.com/materials/polymer_properties/polymer_datasheet.cfm?MajorID=acrylic&MinorID=4

c. For reported low and high values, see: http://www.engineering.com/Library/ArticlesPage/tabid/85/articleType/ArticleView/articleId/152/Thermal-Conductivity.aspx

d. For reported low value, see: http://www.boedeker.com/polyc_p.htm; for reported high value, see: http://en.wikipedia.org/wiki/Polycarbonate
values of the selected surrogate materials other than fused silica are not exactly known. Thus, direct thermal conductivity comparisons of measured values to reported values can only give a perspective of probe accuracy.

5.5.2. Temperature-Dependent Testing

Fused silica was selected for high temperature testing, because the thermal conductivity as a function of temperature is well defined by Touloukian and because it is recommended by ASTM Standard D 5334 - 08 [31]. Temperature-dependent thermal conductivity values for fused silica are shown in Figure 5-12.

![Figure 5-12. Touloukian fused silica recommended thermal conductivity [50].](image-url)
5.6. Needle Probe Transient Method Summary

Using the details of the described testing procedure provided in Section 5.3, thermal conductivity measurements were conducted for the surrogate materials listed in Section 5.5 at room temperature and defined elevated temperatures with results detailed in Chapter 7. Thermal conductivity measurement results were compared to all surrogate material reported values reported in Section 5.5. Sensitivities detailed in Section 5.4 were investigated to determine the accuracy and limitations of the proposed transient method. Sensitivities testing results and discussions are also provided in Chapter 7.
Chapter 6 provides experimentally measured thermal conductivity results for CFOAM25 obtained from the two-thermocouple method. These values are compared with temperature-dependent CFOAM25 thermal conductivity values estimated using material property data (e.g., specific heat capacity, thermal diffusivity, and thermal elongation to estimate density) obtained with HTTL measurement systems. Results from sensitivities analyses listed in Table 4-1 are also detailed in this chapter.

6.1. CFOAM25 Steady-State Two-Thermocouple Experimental Results

Thermal conductivity of the surrogate CFOAM25 material was experimentally measured using the testing procedure described in Section 4.1.3. Two batches of CFOAM25 were tested: the batch 1 sample was used for proof-of-concept validation with constant power setting at 100 watts; and the batch 2 sample was tested for constant power settings of 40 to 100 watts. Batch 1 and 2 results are shown in Figure 6-1, as well as thermal conductivity estimates from properties data results given in Sections 6.2.2 to 6.2.4.

In Figure 6-1, steady-state results over the temperature range of 500 - 600 °C show similar trends with a maximum percentage difference of 14% compared to the thermal conductivity values estimated from properties measurements. Results obtained for the temperature range from 600 - 700 °C, depict a diverging trend with a maximum percentage difference of 33% at approximately 650 °C compared to the thermal conductivity estimated from laboratory material property measurement systems.
Sensitivities from Table 4-1 were investigated. Key results are given in the subsequent subsections.

6.1.1. CFOAM25 Measured Thermal Conductivity
Batch Variation

As noted above, steady-state measurements for the two CFOAM25 batches yielded data that were roughly the same magnitude and reflected similar trends with respect to temperature changes over the 500 to 700 ºC test temperature range. A direct comparison from constant power setting of 100 watts from Batch 1 and 2 in Figure 6-1 show Batch 2 measured results higher in magnitude by roughly 13% around 575 ºC and 21% around 625 ºC, but also show similar trends as temperature increases.

Figure 6-1. CFOAM25 two-thermocouple measured thermal conductivity compared with results estimated from material property testing.
6.1.2. Temperature-Dependent Measured Thermal Conductivity

Figure 6-1 compares results from the two-thermocouple method with experimental data from laboratory systems over the specified temperature range. Between 500 and 600 °C, the maximum percentage difference of values obtained with the two-thermocouple method were within 14% of the estimated average CFOAM25 properties thermal conductivity values obtained from laboratory systems. This percentage difference is in line with the 14% maximum data spread variation from the estimated properties curve (see Section 6.2.4) and the reported 12% measurement uncertainty (see Table 4-2).

CFOAM25 two-thermocouple thermal conductivity measurements diverge from estimated thermal conductivity values for higher temperatures (e.g., between 600 and 700 °C). A maximum percentage difference from the estimated material properties values of 33% occurs at approximately 650 °C. Possible reasons for this diverging data from the two-thermocouple method at higher temperatures are increased natural convection within the porous CFOAM25 sample, and increased radiation heat transfer from the furnace heating coils and the dark-colored CFOAM25 sample.

6.1.3. Constant Supplied Power Sensitivity

Constant supplied power sensitivities data from batch 2 indicate input power had little effect on thermal conductivity measurements. Testing with various supplied power settings, as seen in Figure 6-1, showed data scatter for all constant power settings for temperatures 500 to 600 °C. It does appear, however, that data spread decreases for temperatures ranging from 600 to 700 °C at the constant supplied power settings tested. However, because of the nature of resistance heating, a lower constant supplied power resulted in a
lower temperature drop within the sample. Early investigations determined that supplied power settings of 40 watts or greater provided a temperature drop in the sample sufficient to detect thermal conductivity with reasonable uncertainties.

6.2. HTTL Measured CFOAM25 Property Data

Provided in this subsection are test results for CFOAM25 required material properties for thermal conductivity comparisons of two-thermocouple measurement results, as detailed in Section 4.1.5.3. Specialized systems located at INL's HTTL were used to obtain temperature-dependent thermal conductivity for CFOAM25.

6.2.1. Thermal Diffusivity Measurements

Three CFOAM25 samples from batch 1, with varying thickness (3, 4, and 5 mm), were tested twice to confirm repeatability. From this, it was also deduced that the properties of CFOAM25 change very little when subjected to repeated tests over the testing temperature range of 30 to 1000 °C as test-to-test variations were nearly undetectable. Figure 6-2 shows the average value of batch 1 thermal diffusivity with its upper and lower limits. The upper limits were determined by taking the maximum value as a function of temperature from all tests. Similarly, the lower bound was determined using the same approach.

Three additional CFOAM25 samples from batch 2, with varying thickness were also tested to assess if there were batch-to-batch variations. Batch 1 tests were nearly identical to results from batch 2, and from this it was concluded that batch-to-batch variations were minimal.
6.2.2. Thermal Elongation Measurements for Density Estimates

For density estimates, the thermal elongation of three CFOAM25 samples from batch 1 were tested over a temperature range of 30 to 1000 °C. The density was calculated using the equations and assumptions detailed in Section 4.1.5.3. Samples with different masses and lengths were tested to assess the impact of sample size on thermal expansion data. The results of the dilatometer test are shown in Figure 6-3, where average density is plotted with measurement upper and lower limits.

Figure 6-3 shows that the CFOAM25 density has a linear trend, and that density changes for CFOAM25 are minimal (1.9%) from 100 to 1000 °C. Thermal elongation test-
ing was also very repeatable, as seen by the minimal data spread from the upper and lower bounds in Figure 6-3.

6.2.3. Specific Heat Measurements

Estimates for specific heat capacity of CFOAM25 were completed using three batch 1 machined samples and one batch 1 sample crushed into powder form. As explained in Section 4.1.5.3, closely matching the masses of the reference sample and the unknown sample is recommended for accuracy. CFOAM25 samples machined for measurement in the DSC had masses much lower than the reference sample. However, matching the reference mass was not critical for CFOAM25 estimates because specific heat measurement results from both powder and machined samples were similar in magnitude and trend. Results of calculated average values are shown in Figure 6-4 with upper and lower
bounds. Test temperatures ranged from 30 to 1000 °C. The data spread seen in Figure 6-4 is greater compared to thermal elongation and thermal diffusivity tests, with a maximum percentage difference from the average values of 9.7%. This variation can be attributed to DSC system repeatability for such a porous material.

6.2.4. Thermal Conductivity from Material Properties Data

The temperature-dependent CFOAM25 thermal conductivity was calculated using average values obtained from CFOAM25 material property measurements for density, specific heat, and thermal diffusivity shown in Figures 6-2 through 6-4. CFOAM25 temperature-dependent thermal conductivity with upper and lower bounds is plotted in Figure 6-5. Upper and lower estimates for material properties, which were based on upper and
lower experimental values reported also in Figures 6-2 through 6-4, were less than 14% from the estimated average values with upper values ranging between 8-14% and lower values ranging between 6-12%.

6.3. Two-Thermocouple General Discussion

Two-thermocouple laboratory investigations found that the selected surrogate fuel rod was unable to simulate two desired features that occur in fuel during irradiation, specifically, there were difficulties simulating large temperature gradients within the surrogate rod and distinct temperature-dependent thermal conductivity changes. It is also believed the porosity from the CFOAM25 sample affected high temperature testing due to enhanced internal convection and radiation heat transfer.
However, the two-thermocouple method yielded fairly accurate results for temperatures ranging from 500 to 700 °C. Hence, it is believed that this approach was limited for assessing the effectiveness of the two-thermocouple method. A modified approach, that more closely simulates reactor conditions and different surrogate materials is needed to conduct a more thorough assessment of this approach.
CHAPTER 7
TRANSIENT METHOD RESULTS AND DISCUSSION

Chapter 7 presents needle probe method results for room temperature and elevated temperature testing. Also found in this chapter are results from room temperature and elevated temperature sensitivity measurements with respect to parameters provided in Table 5-5.

7.1. Room Temperature Needle Probe Results

Results given in this section are from room temperature experiments conducted at INL’s HTTL. A room temperature “RT” probe and a high temperature “HT” probe were used to measure room temperature thermal conductivity for five surrogate materials following the testing procedure listed in Section 5.3. Constant power settings ranging from 0.05 to 2 watts were used. Results from all power settings for each surrogate material were averaged and this average measured value is listed in Table 7-1, along with the standard deviation from the average measured value, and percentage difference from the average reported value given from Table 5-7.

Room temperature test results for both fabricated probes compared very well to the ASTM recommended reference material, fused silica, varying less than 1% from the value reported by Touloukian [50]. From this, it was concluded that the correction factor suggested by ASTM 5334-08 (see Section 2.3.3.2) is not needed (probe results were within acceptable limits and the probe diameter was less than the recommended minimum diameter of 2.5 mm).
Sensitivities from Table 5-5 were investigated. Key results are given in the subsequent subsections.

### 7.1.1. Surrogate Material Sensitivity

Results in Table 7-1 indicate that room temperature evaluations from both probes yield accurate data for all surrogate materials tested. All surrogate thermal conductivity results are within the acceptable measurement precision of ±15% stated in ASTM D 5334-08 [31] except measurements from polycarbonate with the RT probe and acrylic from the HT probe. Thus, needle probes designed and fabricated for this thesis research were found

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Material</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average Reported Value (W/m°C)&lt;sup&gt;a&lt;/sup&gt;</td>
<td>SiO₂</td>
</tr>
<tr>
<td>RT Probe Average Conductivity (W/m°C)</td>
<td>1.37</td>
</tr>
<tr>
<td>RT Probe Standard Deviation from Average</td>
<td>0.04</td>
</tr>
<tr>
<td>RT Probe Percentage Difference from Average Reported Value</td>
<td>0.00</td>
</tr>
<tr>
<td>HT Probe Average Conductivity (W/m°C)</td>
<td>1.36</td>
</tr>
<tr>
<td>HT Probe Standard Deviation from Average</td>
<td>0.07</td>
</tr>
<tr>
<td>HT Probe Percentage Difference from Average Reported Value</td>
<td>0.73</td>
</tr>
</tbody>
</table>

<sup>a</sup> See Table 5-7 for surrogate material reported thermal conductivity values.
to detect room temperature thermal conductivity with acceptable accuracies for materials with thermal conductivities ranging from 0.17 to 1.37 W/m\(\cdot\)C.

7.1.2. Supplied Power Sensitivity

Average thermal conductivity values calculated for constant power settings (e.g., 0.05 to 2 watts) were used to generate the results shown in Table 7-1. Room temperature results obtained with higher power settings yielded similar accuracies compared to results obtained from lower power settings. Figure 7-1 shows supplied heater power versus thermal conductivity for the HT needle probe room temperature data. The standard deviation from the average value for each surrogate material measurement demonstrates that variations from the average are small, indicating that the selected magnitude for supplied power has little impact on overall results. This effect is clearly seen in Figure 7-1, as data lie close to the average thermal conductivity line, irrespective of the selected supplied power. However, as stated in Section 2.3.3.5, the power setting must be limited to values that preclude the temperature rise in 1000 seconds from exceeding 10 \(^\circ\)C. Results from these evaluations found this temperature rise criterion more of a general guideline and less critical than stated in ASTM D 5334-08 [31].

7.1.3. Contact Resistance

Gap conductance or a contact resistance plays a critical part in measurement accuracy as stated by Wiesenack and Tverberg [1] and Betten [16]. Most references suggest, if possible, applying a thermal grease or some other medium to enhance the heat transfer between the sample and probe to minimize gap resistance. HTTL tests found that accuracies were better if thermal grease was applied, and errors without thermal grease were
often larger than the acceptable measurement precision of ±15% given in ASTM D 5334-08 [31]. However, thermal conductivity results were sufficiently accurate (most values were within 15% of reported values) if there was a tight fit between the sample and probe without thermal grease.

7.1.4. Slope Calculation Sensitivity

As discussed in Section 5.4, a conservative 5% uncertainty for slope calculation was chosen. For these evaluations, the time-temperature data were plotted on a logarithmic time scale and a trend line was fit to the linear region portion of the data. Experimental data indicate higher power settings (e.g., > 0.5 watt) resolved the temperature as a function of the natural logarithm of time plot better than lower power settings (e.g., < 0.5 watt).
Fluctuations of temperature with respect to time were less with higher power setting, which made slope calculations less susceptible to error. All needle probe data used in these evaluations are provided in the Appendix.

7.2. Temperature-Dependent Testing

Results from temperature-dependent tests are given in this section with results from sensitivity evaluations to the test parameters listed in Table 5-5. The SiO₂ sample was used as the surrogate material in the furnace at 250 and 400 ºC. Constant power settings were varied from 0.25 to 2 watts to assess the HT probe’s ability to detect the thermal conductivity at various power settings. The reported Touloukian value, average calculated thermal conductivity from test results, standard deviation from the average, and percentage difference from the reported value are given in Table 7-2. These HT probe results for the measured thermal conductivity at room temperature, 250, and 400 ºC, are compared in Figure 7-2. Some scatter in the data may be seen at each test temperature; however, the scatter is generally centered around values recommended by Touloukian [50].

Table 7-2. HT Probe Temperature-Dependent Fused Silica Results

<table>
<thead>
<tr>
<th>Temperature (ºC)</th>
<th>Touloukian Reported Value (W/mºC)</th>
<th>Average Measured Conductivity (W/mºC)</th>
<th>Standard Deviation from Average</th>
<th>Percentage Difference from Average Reported Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>1.37</td>
<td>1.36</td>
<td>0.07</td>
<td>0.73</td>
</tr>
<tr>
<td>250</td>
<td>1.65</td>
<td>1.66</td>
<td>0.04</td>
<td>0.61</td>
</tr>
<tr>
<td>400</td>
<td>1.87</td>
<td>1.89</td>
<td>0.04</td>
<td>1.07</td>
</tr>
</tbody>
</table>
Sensitivities to the testing parameters listed in Table 5-5 are discussed with key results given in the subsequent subsections.

7.2.1. Surrogate Material Sensitivity

Results given in Table 7-2 and shown in Figure 7-2 indicate that the HT probe performed well for the reference SiO₂ material at elevated temperatures. The accuracies were less than 2% for all temperatures tested indicating three important results. First, the small standard deviation given in Table 7-2 indicates that the HT probe is repeatable and consistent. Second, the HT probe can detect temperature-dependent changes in thermal conductivity. This is important for in-pile thermal conductivity sensors where the ability to detect temperature-dependent thermal conductivity is critical. Third, data obtained with the HT probe temperature-dependent fused silica results.

Figure 7-2. HT probe temperature-dependent fused silica results.
probe for fused silica at high temperatures is within 2% of the values recommended by Touloukian [50].

7.2.2. Supplied Power Sensitivity

Power settings for elevated temperature testing ranged from 0.25 to 2 watts. The HT probe yielded precise and accurate temperature-dependent measurements at various supplied power settings. Measured data were centered around the recommended Touloukian values at each power setting, shown in Figure 7-3, with the standard deviation slightly greater than 5% (see Table 7-2). Hence, the impact of the selected values for the supplied power setting is minimal.

![Figure 7-3. HT probe temperature-dependent thermal conductivity sensitivity to heater power setting.](image)
7.2.3. Slope Calculation Sensitivity

As ambient temperatures increase, the temperature-dependent results may be affected by slope calculation uncertainties. For the same constant supplied power setting, the measured slope from which thermal conductivity is estimated (see Equation (5.7)) will decrease for samples having larger thermal conductivities. This effect was seen from elevated temperature testing with the HT probe. In effect, the overall temperature rise magnitude in the sample decreases as furnace temperatures increase for the same supplied power. Thus, more supplied power is required as furnace temperature increases to avoid slope calculation errors from temperature fluctuations.

In addition, heat build-up in materials having larger thermal conductivities is not as large as low conductivity materials because conduction heat transfer is more efficient with larger thermal conductivity.

7.3. Summary

Results obtained with the needle probe method were repeatable, consistent, and accurate for the surrogate materials tested, especially for the ASTM reference material, fused silica, at room and elevated temperatures. Results from this research indicate that this transient method offers a superior method for in-pile thermal conductivity detection. The benefits of the needle probe method compared to steady-state two-thermocouple methods include:

- Only heat transfer from the probe to the fuel must be considered, thus eliminating uncertainties from estimating heat transfer from the fuel to the cladding or from the cladding to the coolant, as required for two-thermocouple methods.
• A small, single centerline sensor reduces sample impact; thus point heat source assumptions are better approximated.

• Measurement durations are smaller (e.g., only seconds or minutes for complete transient tests compared to hours or days for steady-state tests, see Sections 2.2 and 2.3 for typical measurement durations).
CHAPTER 8

CONCLUSIONS AND FURTHER CONSIDERATIONS

Two methods for in-pile detection of fuel rod thermal conductivity were investigated using surrogate rod materials in a laboratory setting. A steady-state and a transient method were explored to measure surrogate material thermal conductivity over temperatures ranging from room temperature to 700 °C. Conclusions from experimental findings are given in this chapter, as well as considerations for additional testing.

8.1. Steady-State Two-Thermocouple Method Conclusions

Evaluations were performed to investigate a steady-state two-thermocouple method to assess its viability for detecting in-pile thermal conductivity. Evaluations were completed using a surrogate rod in laboratory tests. Key results from these evaluations include:

- The thermal conductivity of the CFOAM25 rod was measured experimentally using the steady-state two-thermocouple method. Values calculated for constant supplied powers that ranged from 40 to 100 watts and for temperatures that ranged from 500 to 700 °C were found to be within 33% of the average thermal conductivity values obtained from standard material property measurement systems. In addition, values obtained from the two-thermocouple method were consistent with the values obtained from standard property measurement systems (see Figure 6-1) for the specified temperature range. Specifically, values exhibited similar trends and were typically within the upper and lower bounds obtained with laboratory material property measurement systems. Hence, results indicate that the two-thermo-
couple method can detect temperature-dependent thermal conductivity with reasonable accuracies over the temperature range of 500 to 700 °C.

- Sensitivity to measurement parameters listed in Table 4-1 were also investigated. Variations in the constant power supplied to the surrogate sample had minimal impact on measured thermal conductivity results. In addition, no batch-to-batch variations in thermal conductivity values were detected in values obtained from either two-thermocouple measurements or values obtained using laboratory material property measurement systems.

- Temperature-dependent thermophysical properties of elongation, specific heat, and thermal diffusivity, were measured using material property measurement systems to obtain temperature-dependent thermal conductivity of the selected surrogate rod material, CFOAM25. An average thermal conductivity value for CFOAM25 thermal conductivity was calculated using average values from density, specific heat, and diffusivity estimates from several tests. Upper and lower bounds, which bound the data spread from properties measurement repeated testing, were less than 14%.

8.2. Two-Thermocouple Steady-State Method Additional Considerations

As noted in Chapter 6, higher temperature two-thermocouple method evaluations were adversely affected by the experimental setup and the selected surrogate rod material. It should be noted that there were limited materials available to satisfy the test material requirements for a two-thermocouple surrogate material (see Section 4.1.5). The selected surrogate material, CFOAM25, yielded sufficient results over the defined temperature
range (500 to 700 °C); however, results indicate that the porosity of CFOAM25 adversely impacted higher temperature results, precluding the formation of large temperature gradients within the sample.

Suggestions for further study of the two-thermocouple method include: modifications to the experimental setup, alternate surrogate materials, and additional measurement sensitivity testing. Possible method modifications include a centerline heat source to simulate volumetric heat generation instead of Joule heating and/or using a surface heater to enhance and better control the temperature gradient within the sample. Alternate surrogate materials might expand the range of temperatures for method evaluation. However, at this time, no alternative surrogate materials are known that have well-characterized thermophysical properties and satisfy the requirements listed in Section 4.1.5. Additional sensitivities testing could include expanding the temperature range to above 700 °C and below 500 °C with alternate surrogate materials, and use of a modified two-thermocouple approach that considers the effects of fuel-to-cladding heat transfer.

8.3. Transient Needle Probe Method Conclusions

Evaluations were performed to investigate the needle probe method as a potential in-pile technique. Key results from these evaluations include:

- Room and high temperature needle probes were designed and fabricated at INL’s HTTL that incorporated ASTM D 5334-08 [31] recommendations, as well as optimized materials for in-pile applications.
- The room temperature thermal conductivity of five surrogate fuel rods were evaluated using the setup described in Section 5.3.1 and compared to reported values.
Room temperature results listed in Table 7-1 indicate that RT and HT probes accuracies were within 2% for the well-characterized ASTM suggested reference material, SiO₂, and around or under acceptable accuracies of ±15% stated in ASTM D 5334-08 [31] for all surrogate materials.

- Room temperature measurement sensitivity investigations found that supplied constant power settings from 0.05 to 2 watts yielded approximately the same values for thermal conductivity and that the transient method is not greatly impacted by selected input powers. Also as expected, minimizing contact resistance when thermal grease was applied in between the probe and the hole in the sample containing the probe was found to enhance accuracies.

- The setup detailed in Section 5.3.2 was used to experimentally measure fused silica thermal conductivity for temperatures between 250 and 400 °C with constant applied power ranging from 0.25 to 2 watts. Results from these tests were also within 2% of the recommended Touloukian [50] temperature-dependent values.

- Investigations were completed to examine sensitivities of the needle probe method at elevated temperatures to supplied constant power. Results indicate that variations in constant supplied power to the probe did not impact measured thermal conductivity values for fused silica. Also, results indicate that the HT probe can accurately and consistently measure the thermal conductivity of fused silica from room temperature to 400 °C.

Needle probe method test results indicate that surrogate fuel rods can be used in a laboratory setting to gain insights about this approach for in-pile thermal conductivity measurements. Laboratory results using surrogate fuel rod materials suggest that the needle
probe method offers advantages over the steady-state approach. The transient approach minimizes the time required temperature response measurements and reduces the need for assumptions related to heat transfer from the fuel to the cladding or coolant.

8.4. Needle Probe Transient Method Additional Considerations

Several additional investigations are needed to fully quantify the viability the needle probe method for in-pile application, such as maximum measurable thermal conductivity, geometry limitations, upper temperature limit, and long duration performance.

Suggested evaluations should consider selecting materials with higher thermal conductivity values (e.g., k > 4 W/m°C). With higher sample conductivity values, the time required to effectively monitor the temperature response of the sample material is greatly reduced compared to lower sample thermal conductivity values. Thus, efforts to minimize probe response time should be considered from reduced probe diameter and high thermal conductivity probe component materials (e.g., sheath and insulation materials).

Sample and probe geometry effects should also be investigated using typical fuel stack geometries and smaller probe diameters.

As discussed in Section 7.2.3, accurate slope calculations are dependent on avoiding large fluctuations of temperature with respect to the natural logarithm of time. An upper temperature should exist where accurate calculations of the slope are not achievable without exceeding the constant power recommendations provided in Section 2.3.3. Laboratory evaluations are needed to quantify this limit.
Last, typical irradiations in MTRs will last from one to three years. The long duration performance of a representative needle probe at elevated temperature should be evaluated in a laboratory furnace.
REFERENCES


APPENDIX
Appendix A: Needle Probe Temperature Versus Natural Logarithm of Time Plots

All relevant data used in calculations from the needle probe method for RT and HT probes are found in Appendix A. Note all heating and cooling data for a constant power setting are plotting together on a semi logarithmic scale. Cooling data represents the temperature response when constant power to the probe is shut off. Heating data are increasing and cooling data are decreasing with time.

A.1. RT and HT Probe Acrylic Plots From Room Temperature Data

Figure A-1. RT probe acrylic heating and cooling data at constant power of 0.1 watt.
Figure A-2. RT probe arcylic heating and cooling data at constant power of 0.25 watt.

Figure A-3. RT probe arcylic heating and cooling data at constant power of 0.5 watt.
Figure A-4. HT probe acrylic heating and cooling data at constant power of 0.1 watt.

Figure A-5. HT probe acrylic heating and cooling data at constant power of 0.25 watt.
Figure A-6. HT probe acrylic heating and cooling data at constant power of 0.5 watt.
A.2. RT and HT Probe Delrin Plots From Room Temperature Data

Figure A-7. RT probe delrin heating and cooling data at constant power of 0.05 watt.

Figure A-8. RT probe delrin heating and cooling data at constant power of 0.1 watt.
Figure A-9. RT probe delrin heating and cooling data at constant power of 0.25 watt.

Figure A-10. HT probe delrin heating and cooling data at constant power of 0.1 watt.
Figure A-11. HT probe delrin heating and cooling data at constant power of 0.25 watt.

Figure A-12. HT probe delrin heating and cooling data at constant power of 0.5 watt.
Figure A-13. HT probe delrin heating and cooling data at constant power of 1 watt.
A.3. RT and HT Probe Polycarbonate Plots From Room Temperature Data

Figure A-14. RT probe polycarbonate heating and cooling data at constant power of 0.1 watt.

Figure A-15. RT probe polycarbonate heating and cooling data at constant power of 0.25 watt.
Figure A-16. RT probe polycarbonate heating and cooling data at constant power of 0.5 watt.

Figure A-17. RT probe polycarbonate heating and cooling data at constant power of 1 watt.
Figure A-18. HT probe polycarbonate heating and cooling data at constant power of 0.1 watt.

Figure A-19. HT probe polycarbonate heating and cooling data at constant power of 0.25 watt.
Figure A-20. HT probe polycarbonate heating and cooling data at constant power of 0.5 watt.

Figure A-21. HT probe polycarbonate heating and cooling data at constant power of 1 watt.
A.4. RT and HT Probe Particle Board Plots From Room Temperature Data

Figure A-22. RT probe particle board heating and cooling data at constant power of 0.1 watt.

Figure A-23. RT probe particle board heating and cooling data at constant power of 0.25 watt.
Figure A-24. RT probe particle board heating and cooling data at constant power of 1 watt.

Figure A-25. HT probe particle board heating and cooling data at constant power of 0.1 watt.
Figure A-26. HT probe particle board heating and cooling data at constant power of 0.25 watt.

Figure A-27. HT probe particle board heating and cooling data at constant power of 0.5 watt.
Figure A-28. HT probe particle board heating and cooling data at constant power of 1 watt.
A.5. RT and HT Probe SiO$_2$ Plots From Room Temperature Data

Figure A-29. RT probe SiO$_2$ heating and cooling data at constant power of 0.5 watt.

Figure A-30. RT probe SiO$_2$ heating and cooling data at constant power of 1 watt.
Figure A-31. RT probe SiO₂ heating and cooling data at constant power of 2 watts.

Figure A-32. HT probe SiO₂ heating and cooling data at constant power of 0.25 watt.
Figure A-33. HT probe SiO₂ heating and cooling data at constant power of 0.5 watt.

Figure A-34. HT probe SiO₂ heating and cooling data at constant power of 1 watt.
Figure A-35. HT probe SiO$_2$ heating and cooling data at constant power of 1.25 watts.

Figure A-36. HT probe SiO$_2$ heating and cooling data at constant power of 2 watts.
A.6. HT Probe SiO\textsubscript{2} Plots From 250 °C Data

![Figure A-37](image1.png)

Figure A-37. HT probe SiO\textsubscript{2} heating and cooling data at constant power of 0.25 watt.

![Figure A-38](image2.png)

Figure A-38. HT probe SiO\textsubscript{2} heating and cooling data at constant power of 0.5 watt.
Figure A-39. HT probe SiO$_2$ heating and cooling data at constant power of 1 watt.

Figure A-40. HT probe SiO$_2$ heating and cooling data at constant power of 1.25 watts.
Figure A-41. HT probe SiO$_2$ heating and cooling data at constant power of 2 watts.
A.7. HT Probe SiO$_2$ Plots From 400 °C Data

Figure A-42. HT probe SiO$_2$ heating and cooling data at constant power of 0.25 watt.

Figure A-43. HT probe SiO$_2$ heating and cooling data at constant power of 0.5 watt.
Figure A-44. HT probe SiO$_2$ heating and cooling data at constant power of 1 watt.

Figure A-45. HT probe SiO$_2$ heating and cooling data at constant power of 1.25 watts.
Figure A-46. HT probe SiO$_2$ heating and cooling data at constant power of 2 watts.