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In-Pile Thermal Conductivity Measurement Method for Nuclear Fuels

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ABSTRACT

Thermophysical properties of advanced nuclear fuels and materials during irradiation must be known prior to their use in existing, advanced, or next generation reactors. Thermal conductivity is one of the most important properties for predicting fuel and material performance. A joint Utah State University (USU) / Idaho National Laboratory (INL) project, which is being conducted with assistance from the Institute for Energy Technology at the Norway Halden Reactor Project, is investigating in-pile fuel thermal conductivity measurement methods. This paper focuses on one of these methods – a multiple thermocouple method. This two-thermocouple method uses a surrogate fuel rod with Joule heating to simulate volumetric heat generation to gain insights about in-pile detection of thermal conductivity. Preliminary results indicated that this method can measure thermal conductivity over a specific temperature range.

This paper reports the thermal conductivity values obtained by this technique and compares these values with thermal property data obtained from standard thermal property measurement techniques available at INL's High Test Temperature Laboratory. Experimental results and material properties data are also compared to finite element analysis results.

KEYWORDS: In-pile instrumentation, nuclear fuel thermal conductivity

INTRODUCTION

Thermophysical properties must be known prior to deploying new fuels and structural properties in nuclear reactors. Thermal conductivity, one of the most important properties for predicting fuel and material performance, is highly dependent on physical structure, chemical composition, and state [1]. The physical structure and chemical composition of nuclear fuels change during irradiation as a function of time and position within the rod. At the Idaho National Laboratory's (INL's) Advanced Test Reactor (ATR), most thermophysical properties, including thermal conductivity degradation, are measured out-of-pile in "hot-cells." Samples are irradiated for a specific period of time and removed for testing. This testing has several disadvantages. It is expensive to repeatedly remove samples from the reactor, examine them out-of-pile, and return them to the reactor. Furthermore, this process may disturb phenomena of interest, and only provides a view of the sample's end state at the time each measurement is made.

Few in-pile thermal conductivity measurement techniques currently exist, and existing techniques invoke numerous assumptions to obtain data. The Institute for Energy Technology (IFE) at Norway's Halden Reactor Project (HRP) has applied a method similar to the two thermocouple approach to detect thermal conductivity degradation during irradiation [2]. IFE HRP calculates thermal conductivity degradation as a function of fuel rod burnup from by fuel centerline temperatures obtained from an embedded thermocouple combined with well known heat flux and thermal hydraulic conditions [3]. Although not explicitly stated in References [2], [3], and [4], this approach must assume several conditions about the fuel, such as uniform fuel composition, uniform fuel density, gap conductances [4], and uniform heat generation distribution of the fuel rod. It is the aim of this USU/INL research to better understand these assumptions.

USU/INL evaluations, with assistance from researchers at the IFE HRP, calculate thermal conductivity using two thermocouples inserted into a surrogate fuel rod, one to monitor fuel centerline temperature and another to monitor temperature at a measured radial position within the rod.

Although not discussed in this paper, USU/INL investigations will ultimately consider the use of hot wire [5, 6] methods to directly detect changes in fuel thermal conductivity. Preliminary investigations [7] indicate that this approach may offer advantages over two-thermocouple techniques.

APPROACH

The USU/INL two-thermocouple method is based in a laboratory setting using a surrogate material, and is conducted at INL's High Temperature Test Laboratory (HTTL). The research includes three components for method validation: first, experimental thermal conductivity readings of the proposed two-thermocouple method; second, thermophysical properties measurements to calculate the temperature-dependent thermal conductivity of the surrogate material; and third, Finite Element Analysis (FEA) to explore gap conductance sensitivities. This section describes the approach used for each component of this research.

Surrogate Material Selection and Characterization

The surrogate material allows for method validation testing in a safe, cost effective environment while providing essential insights in preparation for *in-situ* testing. The chosen surrogate fuel rod material for this proof-of-concept test is CFOAM25 manufactured by Touchstone Research Laboratory [8]. Important material property selection criteria of the surrogate material were: electrical resistivity, thermal conductivity, and temperature limit in air and inert test conditions. Few materials matched needed properties, and CFOAM25 appeared the best choice. Data from Touchstone were useful for preliminary selection of CFOAM25; however, more detailed, temperature-dependent, material property data were needed for the USU/INL evaluations. Temperature-dependent data obtained using standard material property measurement systems (e.g., laser flash diffusivity, pushrod dilatometry, and differential scanning calorimetry) available at INL's High Temperature Test Laboratory (HTTL). As documented in Reference [9], CFOAM25 temperature-dependent thermal conductivity was estimated using Equation (1) and temperature-dependent thermal diffusivity, α ; density, ρ ; and specific heat capacity, C_p data:

$$k = \alpha \cdot \rho \cdot C_p. \quad (1)$$

Upper and lower estimates for CFOAM25 material properties, which were based on data scattering from properties testing, were no greater than 14% from the estimated average values with upper values ranging between 8%-14% and lower values ranging between 6%-12%.

Two-thermocouple Method Theory and Setup

The method uses the well-defined principle of radial heat flow. The method for quantifying the steady state thermal conductivity of a fuel rod, k , can be obtained from two-thermocouple method by starting with Fourier's Law in cylindrical coordinates. The thermal conductivity of a rod at any radial position can be estimated from [9]:

$$k(r) = \frac{\dot{q} \cdot r^2}{4 \cdot \Delta T}, \quad (2)$$

Hence, thermal conductivity can be calculated if the radial position from the sample centerline, r ; volumetric heat generation, \dot{q} ; and measured temperature difference, ΔT , are precisely known. The test setup shown in Figure 1 is being used to obtain data for these parameters.

Figure 1 shows a tube furnace used only to control ambient conditions, a power supply to provide Joule heating within the sample, a shunt to measure current, Type K thermocouples to measure sample temperatures at the centerline and 3/8" away from the centerline (as seen in Figure 2), and a data acquisition system to record signals from these measurements.

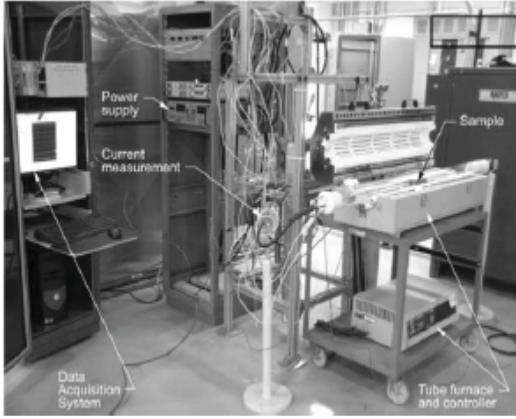


Figure 1. Experimental test setup at INL's HTTL

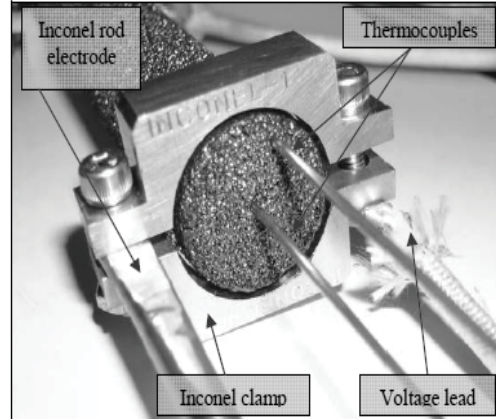


Figure 2. CFOAM sample setup

Figure 2 shows the CFOAM sample connected to the current loop using Inconel electrodes. The heat generation rate can be calculated using the sample geometry (measured before testing), and real time measurements of current and voltage. Two holes machined in the sample, allow for the 1/16" Type K thermocouples to be inserted so that the thermocouple junction is located 3" of the 6" sample length. The environment in the tube can be controlled by varying flow rates of air or argon.

Sensitivities tests were conducted to view the impact of constant power level, temperature gradient, gas flow, and furnace temperature.

There are acknowledged limitations to the two-thermocouple method. Placing two thermocouples within a prototypic-sized 1/2" diameter fuel rod will incur significant perturbations in the measured fuel thermal conductivity. Another limitation of the method, which is inherent to measuring temperature with thermocouples, is the contact resistance between the thermocouples and sample.

As documented in Reference [9], the remaining experimental measurement uncertainties were quantified by applying the approach suggested in Reference [13] to Equation (2). Substituting $\dot{q} = (I*V) / ((\pi*r_o)^2*L)$, where V is the measured sample voltage drop, and I is the measured current. The partial derivatives of each measured parameter were obtained to approximate the uncertainty from the two-thermocouple method as:

$$\varepsilon_k = \sqrt{(\varepsilon_V)^2 + (\varepsilon_I)^2 + 2 \cdot (\varepsilon_r)^2 + 2 \cdot (\varepsilon_{r_o})^2 + (\varepsilon_{\Delta T})^2 + (\varepsilon_L)^2}, \quad (3)$$

where, ε_V is the uncertainty from the power supply voltage given by the manufacture; ε_I is the calibration uncertainty from the shunt; ε_r is the measurement uncertainty between thermocouples; ε_{r_o} is the initial rod radius measurement uncertainty; $\varepsilon_{\Delta T}$ is the temperature uncertainty given by thermocouple manufacture; ε_L is the length measurement uncertainty; and ε_k is the total measurement uncertainty of rod thermal conductivity.

The largest contributing source of error from Equation (3) is from the measured distance between the thermocouples. As noted above, this analysis does not include uncertainties associated with the thermocouples being a different material than the

surrogate rod material or contact resistances. The maximum calculated measurement uncertainty from Equation (3) was found to be just over 12%.

Finite Element Analysis

Finite Element Analyses (FEA) were completed with the Abaqus 6.8-2 [10] code. The Abaqus model was developed to provide insights and comparisons regarding the experimental results and to help bound the potential effects of non-ideal contact thermal resistance.

Abaqus was used to generate a 3-D model of the experimental setup used to measure the steady state thermal conductivity of a fuel rod surrogate. Key features of the model include representations of CFOAM25 rod, 1/16" type K thermocouples, and gap elements used to evaluate the effects of conduction contact resistance.

CFOAM25 material property data were used to define the model thermal parameters as a function of temperature. The model was constructed as a single extruded three-dimensional deformable part, with separate components (surrogate rod material, thermocouples, and gaps) defined by partitions. The gap elements were modeled as solids in order to simplify the model (with effective gap heat transfer coefficient simplified to a conductivity value). A close up view of the assembled model structure is shown in Figure 3.

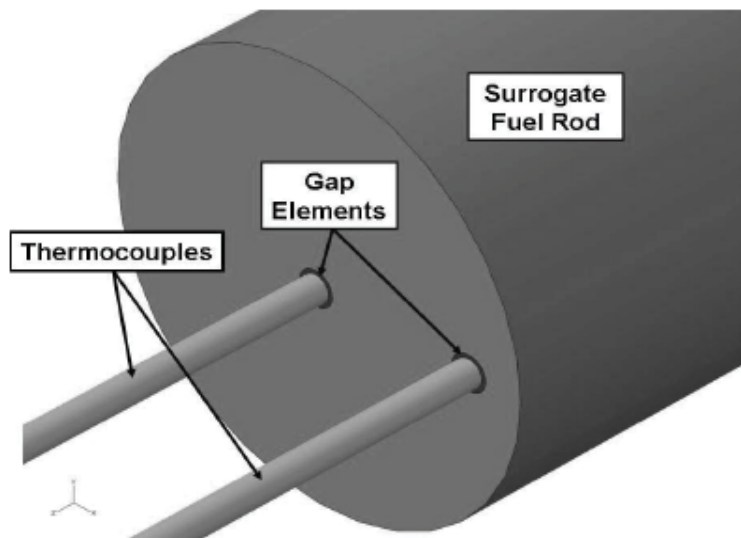


Figure 3. Abaqus model assembly showing thermocouples, gap elements, and surrogate rod

The density and specific heat capacity of the gap were characterized using properties of argon, the gas in which the test was conducted. Thermal conductivity of the gap was derived using Equation (4) [11]:

$$h_{gap} = \frac{k_g}{3.6(R_1 + R_2) + (g_1 + g_2)}. \quad (4)$$

Where:

- k_g - thermal conductivity of argon (W/m²K)
- R_1, R_2 - surface roughness of materials on each side of the gap (m),
this value was estimated for CFOAM25
- g_1, g_2 - temperature jump distances (m)

and

$$g_1 + g_2 = \frac{0.0247k_g T_{gas}^{0.5}}{P_{gas} \frac{a_{gas}}{M_{gas}}}. \quad (5)$$

Where:

- T - temperature of the argon (K), taken as furnace temperature
- P - pressure of the argon (Pa), taken as atmospheric
- M - molecular weight of argon (kg/kg*moles)
- a - accommodation factor [12], ranging between 0 and 2.

The thermal conductivity of the gap elements was then by defined as h_{gap} multiplied by the thickness of the element.

Abaqus allows direct application of volumetric heat loads. However, due to the complexity of estimating both convective and radiation cooling coefficients as functions of temperature, experimental data were used to approximate surface cooling. The film cooling coefficient was adjusted such that the peak model temperature registered by the centerline thermocouple matched closely that given in the experiment.

The experiment was conducted using total power dissipations of 40 and 100 watts. The model was run as a transient analysis, with the end condition determined as steady state (defined as a temperature change of less than 0.001 °C between iterations, a steady state analysis was used as a check). Temperatures recorded at nodes located at the thermocouple junctions were used for determining ΔT and effective thermal conductivity of the simulated CFOAM25.

RESULTS AND DISCUSSION

Experimental Results

Initial studies have focused on a sample temperature range of 500 – 700 °C. While testing is still ongoing to evaluate method sensitivities, results from early testing shows that the method can accurately measure the surrogate rod thermal conductivity within the defined temperature range.

Sample and furnace equilibrium temperature were observed to have a direct relationship to input power through the sample; larger supplied power and higher furnace temperatures yielded higher equilibrium temperatures for obtaining data. In general, data obtained at higher power levels and for lower furnace temperatures yielded larger temperature gradients through the samples. These larger temperature gradients, in turn, yielded thermal conductivity values more consistent with values obtained from material property systems at the HTTL.

Several observations can be made from early results. The experimental values ranged from 2 - 8% of the values in the material properties curve, and were within the 14% uncertainty range of the material properties measurement of the CFOAM25 sample [9]. These experimental readings are also within the measurement uncertainty of 12%. The values are of the same order of magnitude and show similar trends over the defined temperature range. Testing is ongoing at INL's HTTL to fully understand the limitations of this method.

ABAQUS Results

Using CFOAM25 material properties and equations in References [11] and [12], thermocouple to CFOAM25 gap conductances were calculated to range from 17.5 to 3270 W/m²K. Abaqus calculations were completed to assess the sensitivity of gap conductance for outer and centerline thermocouples for power inputs of 40 W and 100 W and ambient temperatures ranging from 400 – 800 °C. Estimates from Abaqus calculations for gap conductance ranges are compared with average material property test results in Figure 4.

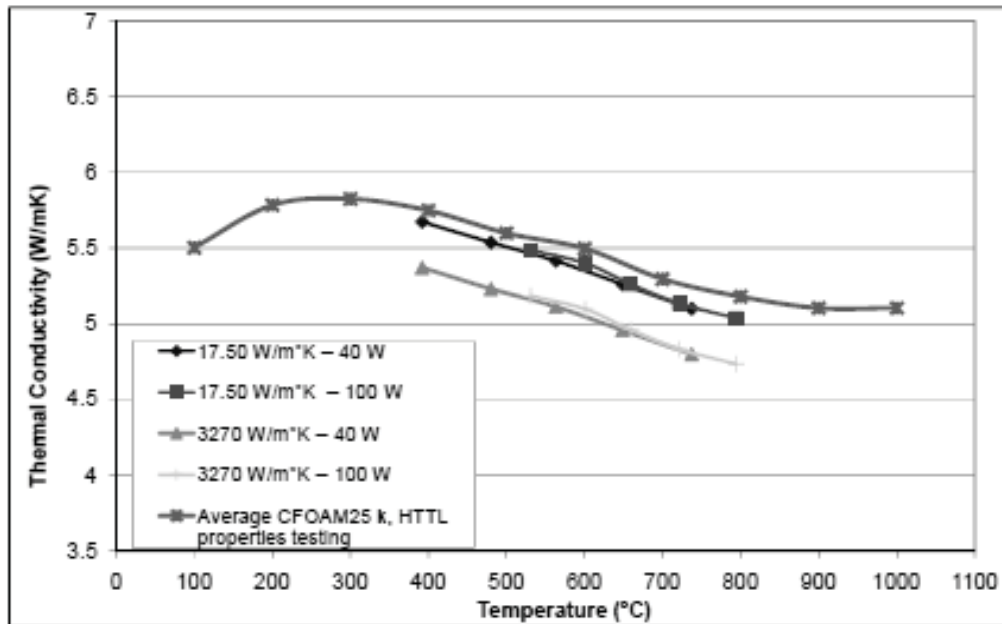


Figure 4. Abaqus results for power inputs of 40 and 100 W

As shown in Figure 4, values derived with Abaqus are consistent with measured CFOAM25 material property values and experimental values. Comparisons reveal similar trends with temperature and estimated values are within or slightly below estimated properties bounds. Perhaps more important, it is worth noting that extreme variations in gap coefficients produce small, variations in output thermal conductivity values. For example, the maximum value for gap coefficient of 3270 W/m²K is approximately 187 times greater than the lowest estimate of 17.5 W/m²K. Although this increase of 187 in gap conductance did change estimates for thermal conductivity, the change was limited to approximately 6%. Hence, variations from gap coefficient selections would indicate that large changes in gap conductance effects have minimal impact on detecting changes in thermal conductivity. This insight is extremely important in cases where in-pile variations in gap conductance during irradiation are difficult to quantify.

CONCLUSIONS

As discussed in this paper, evaluations of in-pile thermal conductivity measuring techniques are first investigating a method which uses two positioned thermocouples to measure the temperature of two points within a surrogate rod. Results of thermal conductivity measurements from this rod are compared to CFOAM25 properties estimated using laboratory systems available at INL's HTTL. Also, results from FEA are used for additional validation and to evaluate the effects of conduction contact resistance.

While limitations to the method are acknowledged, initial results provide key insights about this method as an in-pile measurement approach. The key conclusions from this paper are:

- Experimental results were used to calculate the surrogate rod thermal conductivity in the temperature range of 500 – 700 °C. Initial results showed higher input values of power gave results closer to the properties measurement, and were found to be within 2-8% when input power was 100 watts [9]. Values obtained experimentally are consistent with the values obtained from standard property measurement systems and FEA results over the defined temperature range.
- FEA sensitivity calculations indicate that large changes in gap conductance have minimal impact on detecting changes in thermal conductivity. Hence, these results suggest that uncertainties in gap conductance during irradiation may not impact the ability of the two-thermocouple method to detect thermal conductivity degradation.

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