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Development of an In-Pile Technique for Thermal Conductivity Measurement

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Brandon S. Fox
Heng Ban
Joy Rempe
Joshua Daw
Keith Condie
Darrell Knudson

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DEVELOPMENT OF AN IN-PILE TECHNIQUE FOR THERMAL CONDUCTIVITY MEASUREMENT

Brandon S. Fox and Heng Ban
Utah State University
4130 Old Main Hill
Logan, UT 84322-4130
b.fox@aggiemail.usu.edu; heng.ban@usu.edu

Joy Rempe, Joshua Daw, Keith Condie, and Darrell Knudson
Idaho National Laboratory
785 DOE Place
Idaho Falls, ID 83402
Joy.Rempe@inl.gov; Joshua.Daw@inl.gov; Keith.Condie@inl.gov; Darrell.Knudson@inl.gov

ABSTRACT

Thermophysical properties of advanced fuels and materials during irradiation must be known prior to their use in existing, advanced, or next generation reactors. Fuel thermal conductivity is one of the most important properties for predicting fuel performance and reactor safety. This paper discusses 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 Halden Reactor Project (IFE HRP), to investigate in-pile fuel thermal conductivity measurement methods using a surrogate fuel rod. The methods use a surrogate fuel rod with Joule heating to simulate volumetric heat generation to gain insights about in-pile detection of thermal conductivity. Initial investigations have focused on a carbon structural foam, CFOAM®, a product of Touchtone Research Laboratory as a surrogate material because of its electrical and thermal properties.

This paper describes the method used to evaluate in-pile thermal conductivity measurement methods. A description of the test setup and preliminary results are presented. Thermal conductivity values obtained by this technique are compared with thermal property data obtained from standard thermal property measurement techniques, such as laser flash, differential scanning calorimetry, and pushrod dilatometry.

Key words: Fuel Rod Thermal Conductivity Measurement, In-Pile Instrumentation

1 INTRODUCTION

Thermophysical properties of materials must be known for proper design, test, and application of new fuels and structural properties in nuclear reactors. Because thermal conductivity is a transport property, the diffusion process of energy transfer is greatly dependant on the physical structure, chemical composition, and state of the material [1]. In the case of nuclear fuels during irradiation, the physical structure and chemical composition change during irradiation as a function of time and position within the rod. Typically, thermal conductivity changes, as well as other thermophysical properties are measured out-of-pile in "hot-cells," after these samples are irradiated for a specific length of time. Repeatedly removing samples from the reactor to make out-of-pile measurements is expensive, has the potential to disturb phenomena of interest, and only provide 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 the development of next generation reactors and advanced fuels for existing nuclear plants. Having the capacity to effectively and quickly characterize fuel properties during irradiation has the potential to improve the fidelity of nuclear fuel data and reduce irradiation costs.

Few in-pile measurement techniques for thermophysical properties are currently available. However, the Institute for Energy Technology (IFE) at the Halden Reactor Project (HRP) developed an approach for detecting fuel rod conductivity degradation during irradiation [2]. The approach relies on data from a thermocouple in the fuel rod to give centerline temperature and a second thermocouple placed on the exterior of the fuel cladding. Fuel thermal conductivity was calculated to obtain the plot shown in Figure 1.

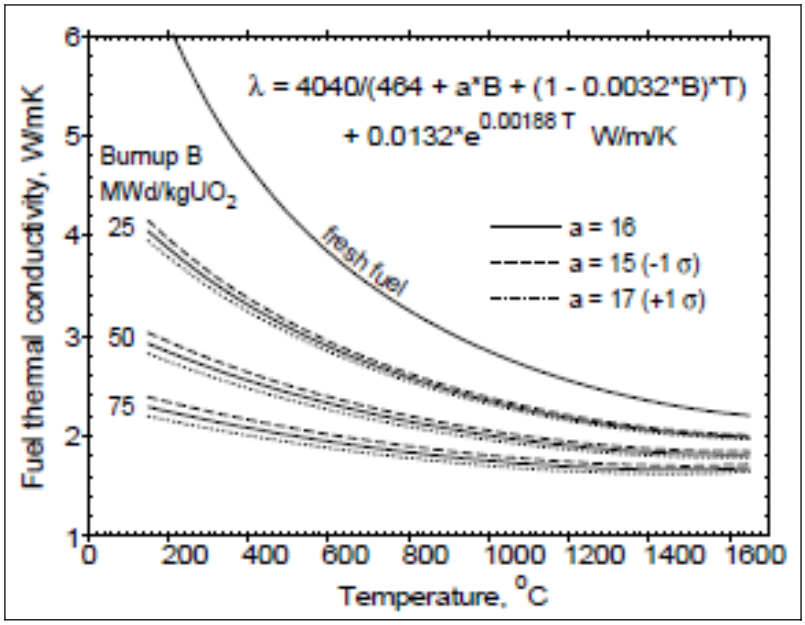

Figure 1. Halden plot of fuel conductivity as a function of rod burn-up [2]

Initial USU/INL evaluations, which are being performed with assistance from researchers at the IFE HRP, calculate fuel rod thermal conductivity 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 technique is being tested under several conditions to view the sensitivity of the measurement. Although initial USU/INL evaluations are performed using 1/16" diameter Type K thermocouples, it is proposed that irradiations incorporate the use of INL-developed High Temperature Irradiation Resistant Thermocouples (HTIR-TCs) whose doped molybdenum/niobium alloy thermoelements do not experience degradation at high temperatures (long duration testing demonstrates their stability up to temperatures of 1500 ºC) or decalibration due to transmutation [3]. Although not discussed in this paper, USU/INL investigations will ultimately consider the use of hot wire methods to directly detect changes in fuel thermal conductivity. Preliminary investigations [4] indicate that this approach may offer advantages over two-thermocouple techniques.

1.1 Surrogate Rod Material

The initial surrogate fuel rod material chosen for this proof-of-concept test is CFOAM®. This carbon structural foam is non-combustible and will not off-gas at high temperatures. CFOAM® is calcined coke (CAS #64743-05-1) [5] engineered to meet high performance material needs. It has a high tolerance to impact damage and can be integrated with other materials, such as metals or polymer composites. Table I from Reference [5] summarizes properties provided by Touchtone Research Laboratories Ltd. for this material. Although these data were useful for preliminary selection of CFOAM®, more detailed, temperature-dependent, material property data are needed for the USU/INL evaluations. This paper documents the measurement 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).

Table I. CFOAM properties

Property	Test Method	CFOAM20	CFOAM25	Unit
Nominal Density	ASTM D1622	20 0.32	25 0.4	lbs/ft^3 g/cc
Thermal Conductivity	ASTM E 1225	0.15 to 16 0.25 to 25 (Can be tailored for specific applications)		BTU/ft-hr°F W/m°K
Maximum Operational Use Temperature		1100 Air 600 Air		°F °C
		5500 Inert gas 3000 Inert gas		
Electrical Resistivity[1]	ASTM D 4496	4E-03 to eE+06 1E-02 to 1E+07 (Can be tailored for specific applications)		Ohm-in Ohm-cm

[1] Data at Room Temperature or 20 ºC.

1.2 CFOAM® Property Measurements

In order to properly validate proposed methods estimating thermal conductivity, properties of surrogate fuel rods must be quantified. Three important property measurements must be made in order to estimate the material's thermal conductivity. These values can be found by standard laboratory equipment located at INL's HTTL. Although two CFOAM® materials (CFOAM20 and CFOAM25) were initially considered, the denser CFOAM25 was found to have more desirable properties for this application. CFOAM25 property values are given in this paper.

1.2.1 Density measurements

The density of a material as a function of temperature can be 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:

$$\alpha_L = \frac{1}{L_o}\frac{\partial L}{\partial T}, \tag{1}$$

where L_o is the initial length. The above expression is often rewritten as:

$$\frac{\Delta L}{L_o} = \alpha_L \cdot \Delta T, \tag{2}$$

with ΔL is the sample change in length, α_L is the coefficient of linear expansion, and ΔT 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:

$$\frac{\Delta V}{V_o} = 3 \cdot \alpha_L \cdot \Delta T, \tag{3}$$

where ΔV is the sample volume change, and V_o 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, sample change in sample length, as shown by equation (4):

$$\rho_f = \frac{m}{V_o(1+3\frac{\Delta L}{L_o})}. \tag{4}$$

Three CFOAM25 samples were tested over a temperature range of 30 ºC to 1000 ºC. Each sample had a different mass and length to view the measurement sensitivity. The results of the dilatometer test are shown in Figure 2, where average density is plotted with measurement upper and lower limits.

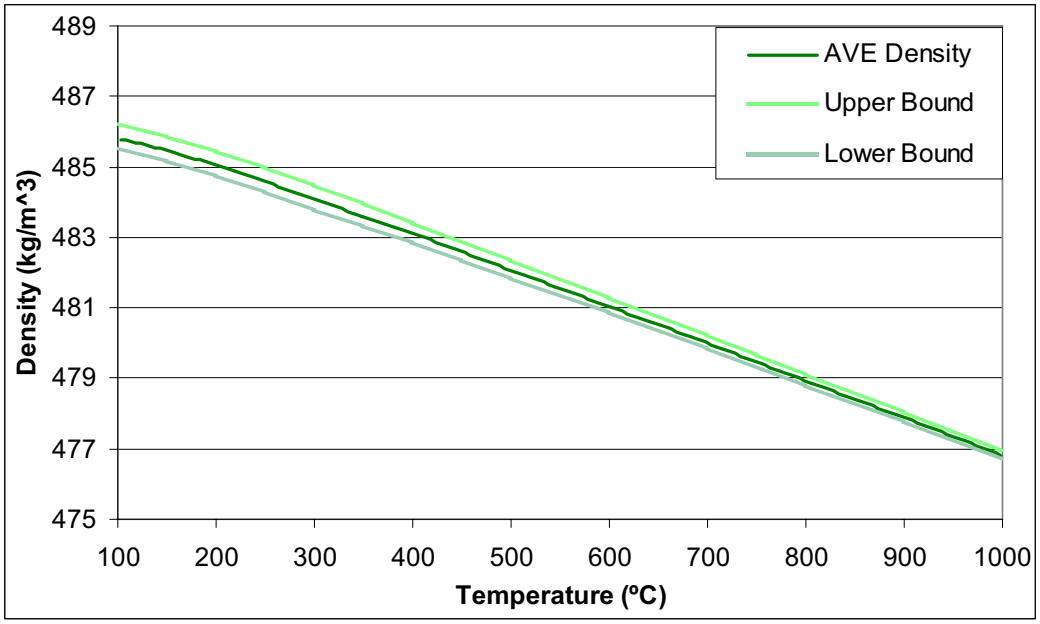

Figure 2. CFOAM® average density vs. temperature with upper and lower bounds

Figure 2 shows that the CFOAM25 density can be approximated linearly and that density changes for CFOAM25 are minimal (1.9%) over the testing temperature range.

1.2.2 Specific heat measurements

Specific heat 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 sample 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. CFOAM25 samples were machined to the required test size geometry, but the sample masses were much lower than the lowest mass of reference samples available. Tests were conducted to view the variation of matching unknown sample and reference masses. Hence, CFOAM25 tests were completed using three machined samples and one sample crushed into powder form. The test temperature ranged from 30 ºC to 1000 ºC. An average was calculated from these tests and results are shown in Figure 3 with upper and lower bounds.

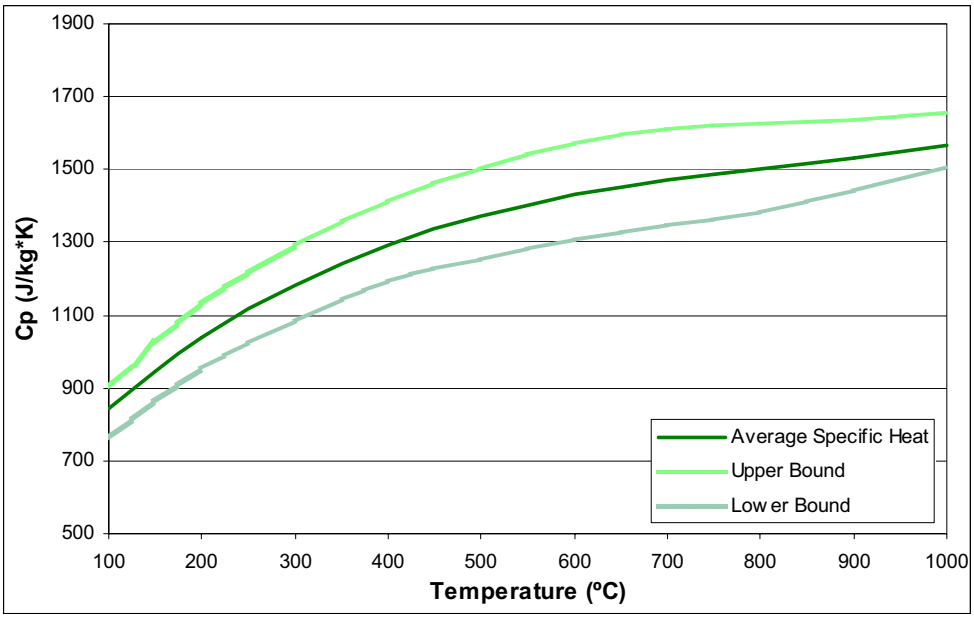

Figure 3. CFOAM® average specific heat, C_p, vs. temperature with upper and lower bounds

It was speculated that the powdered/crushed test is more accurate because the powder mass more closely matched the mass of the reference sample. The machined samples were about half the mass of the reference sample. However, the average value with upper and lower bounds was used in this effort to characterize the material's specific heat capacity.

1.2.3 Thermal diffusivity measurements

Thermal diffusivity, α, 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 [6].

Thermal diffusivity can be measured in a laboratory setting 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. Three CFOAM25 samples, with varying thickness, were tested twice to confirm repeatability. From this, it is also deduced that the properties of CFOAM® change very little when subjected to repeated tests over the testing temperature range of 30 ºC to 1000 ºC as test to test variations were almost undetectable. Figure 4 shows the average value of thermal diffusivity with its upper and lower limits.

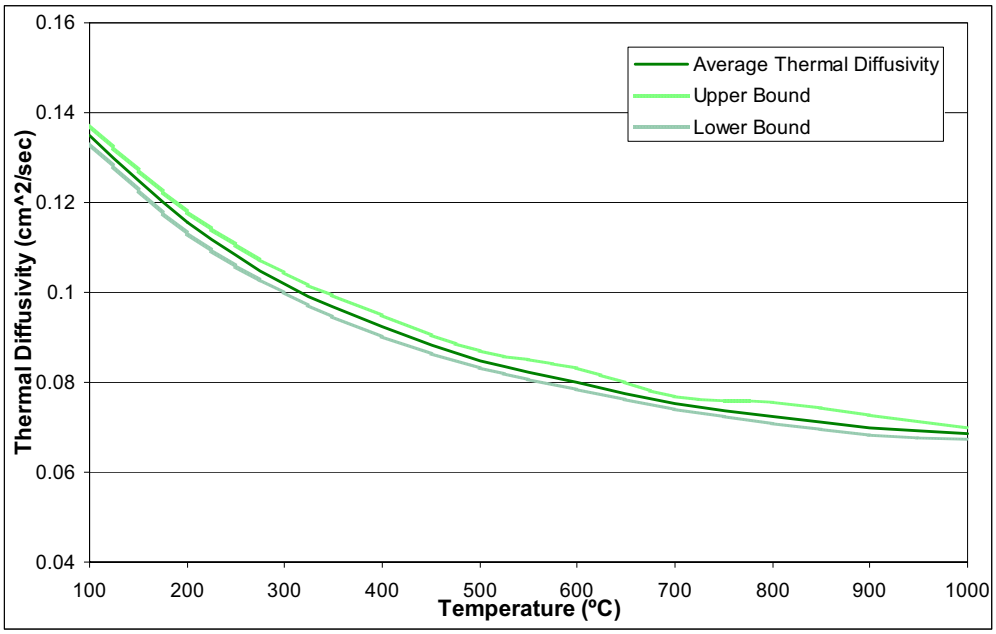

Figure 4. Average diffusivity with upper and lower bounds

1.3 Thermal Conductivity from Property Measurements

As previously mentioned, the thermal conductivity of a material can be related to its thermodynamic properties, density, specific heat, and thermal diffusivity by:

$$k = \alpha \cdot \rho \cdot C_p . \tag{5}$$

The CFOAM25 thermal conductivity is calculated using average values obtained from CFOAM25 material property measurements for density, specific heat, and thermal diffusivity shown in Figures 2 through 4. Upper and lower estimates for material properties, which were based on upper and lower experimental values reported also in Figures 2 - 4, were no greater than 14% from the estimated average values with upper values ranging between 8%-14% and lower values ranging between 6%-12%.

1.4 Thermal Conductivity Basis

As shown in Figure 5, the method for quantifying the steady state thermal conductivity of a fuel rod, k, can be obtained from two-thermocouple technique data using the following parameters:

- $\dot{q}$ - volumetric heat generation rate
- r_o - radius of the rod
- L - rod length
- T_s - rod surface temperature
- T_∞ - ambient air temperature
- h - convective heat transfer coefficient.

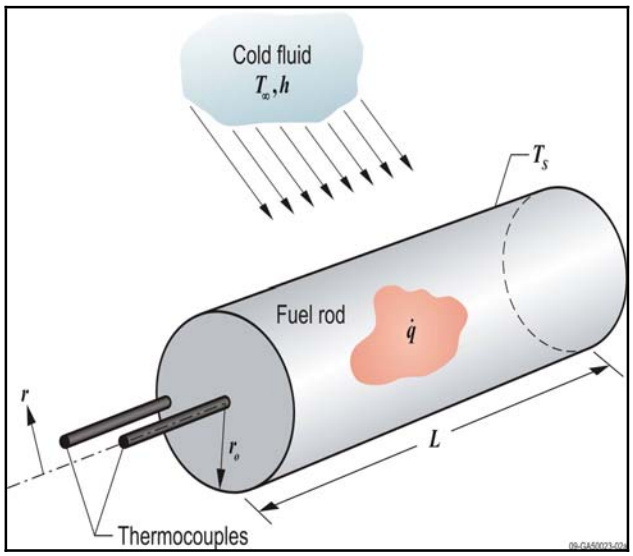

Figure 5. Solid cylinder heat conduction with uniform volumetric heat generation

Starting with the cylindrical form of the heat equation with uniform heat generation, a definition for thermal conductivity can be found:

$$\frac{1}{r}\frac{d}{dr}\left(r\frac{dT}{dr}\right)+\frac{\dot{q}}{k}=0\,. \tag{6}$$

Using separation of variables, applying a zero temperature gradient at the rod centerline, and assuming a surface conduction/convection balance boundary condition, the temperature distribution in the rod as a function of radial position can be defined as:

$$T(r)=\frac{\dot{q}\cdot r_o^2}{4\cdot k}\left(1-\frac{r^2}{r_o^2}\right)+T_s\,. \tag{7}$$

At a distance, r, from the centerline of the rod, the following relationship for thermal conductivity can be obtained:

$$k=\frac{\dot{q}\cdot r^2}{4\cdot \Delta T}\,, \tag{8}$$

where ΔT is the temperature difference between the centerline and the radial position. Hence, thermal conductivity can be calculated if the radial position from the sample centerline, r; heat generation, $\dot{q}$; and measured temperature difference, ΔT, are precisely known.

1.4 Experimental Method

From the above definition of thermal conductivity, the required measured parameters are: distance from centerline to outer thermocouple, temperatures from each thermocouple, and heat generation rate. The test setup shown in Figures 6 and 7 is being used to obtain data for these parameters.

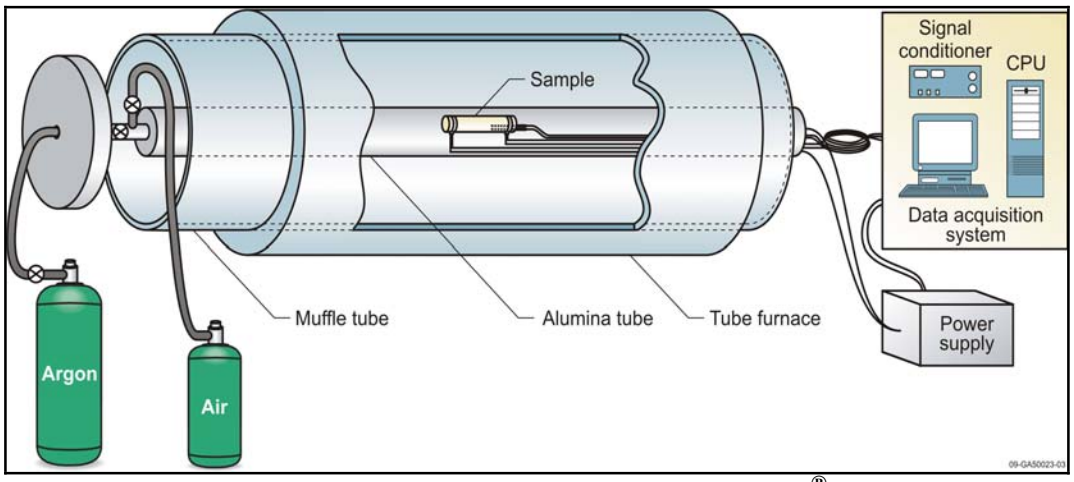

Figure 6. Test setup inside tube furnace CFOAM® sample

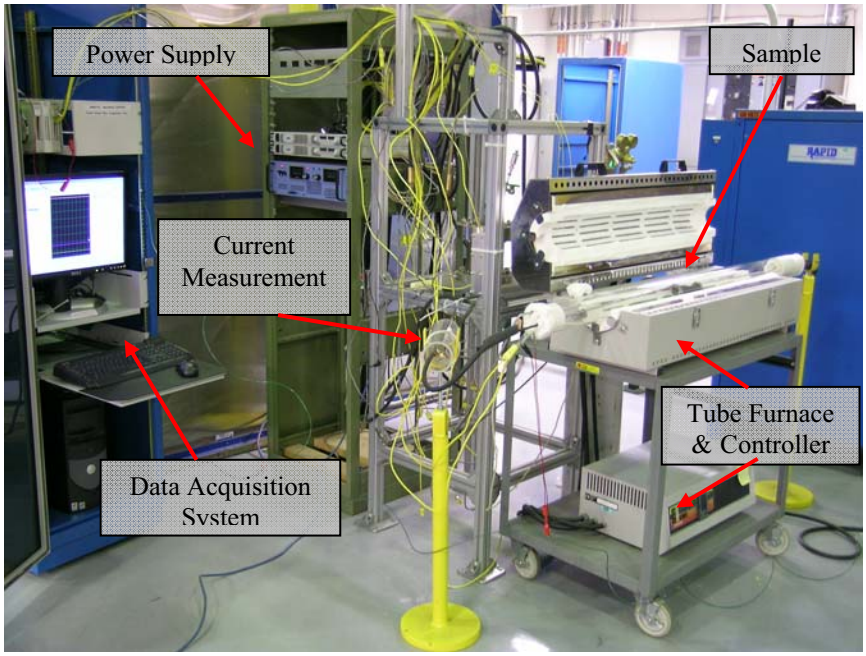

Figure 7. Test setup at INL's HTTL

The samples are positioned inside a tube furnace to control temperature and provide a sample temperature test range from 500 ºC – 700 ºC. The tube furnace only changes ambient temperature variation. A specified voltage and current are supplied to the sample by attaching the power supply to each end of the sample using Inconel electrodes connected to Inconel clamps. Leads attached to Inconel clamps at each end of the surrogate rod measures the voltage drop of the sample. A precision current measurement measures current within the experimental test loop. Volumetric heat generation is calculated using the measured current, I; and the sample voltage drop, V; sample dimensions, and the relationship P = I*V. Flow rates can be adjusted using valves to vary fluid conditions within the tube. The fluid inside the tube can either be air or an inert gas, such as argon. Signals are processed by a data acquisition system to give temperatures from thermocouples and power in the sample. The thermocouples are carefully positioned at known locations within the sample as seen Figure 8:

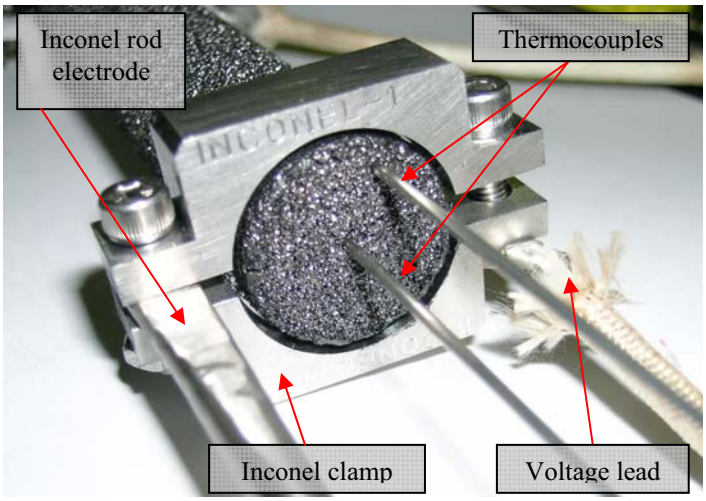

Figure 8. CFOAM20 sample with thermocouples inserted

1.5 Testing Variations to Detect Method Sensitivities

Several types of materials will be considered in USU/INL evaluations. In addition, several test parameters will be varied to estimate the accuracy of the proposed method. Table II shows proposed variations for steady state conditions.

Table II. Testing variations for steady state conditions

Sensitivity	Experimental Parameter Varied
Variation in Temperature	Vary furnace temperature (300 ºC to 500 ºC)
Variation in Temperature Gradient	Vary power supply voltage
Variation in Outer Boundary Condition	Vary fluid flow along outer surface of sample

2 RESULTS

Thermal conductivity testing began at INL's HTTL in January 2009 and is still ongoing. However, testing to date has proven to be very successful with the setup shown in Figure 7. Testing was focused within the 500 ºC – 700 ºC temperature range. Selected results from early testing with the supplied power held constant at 100W are compared in Figure 9 with average thermal conductivity values obtained from material property measurements shown in Figures 2 through 4.

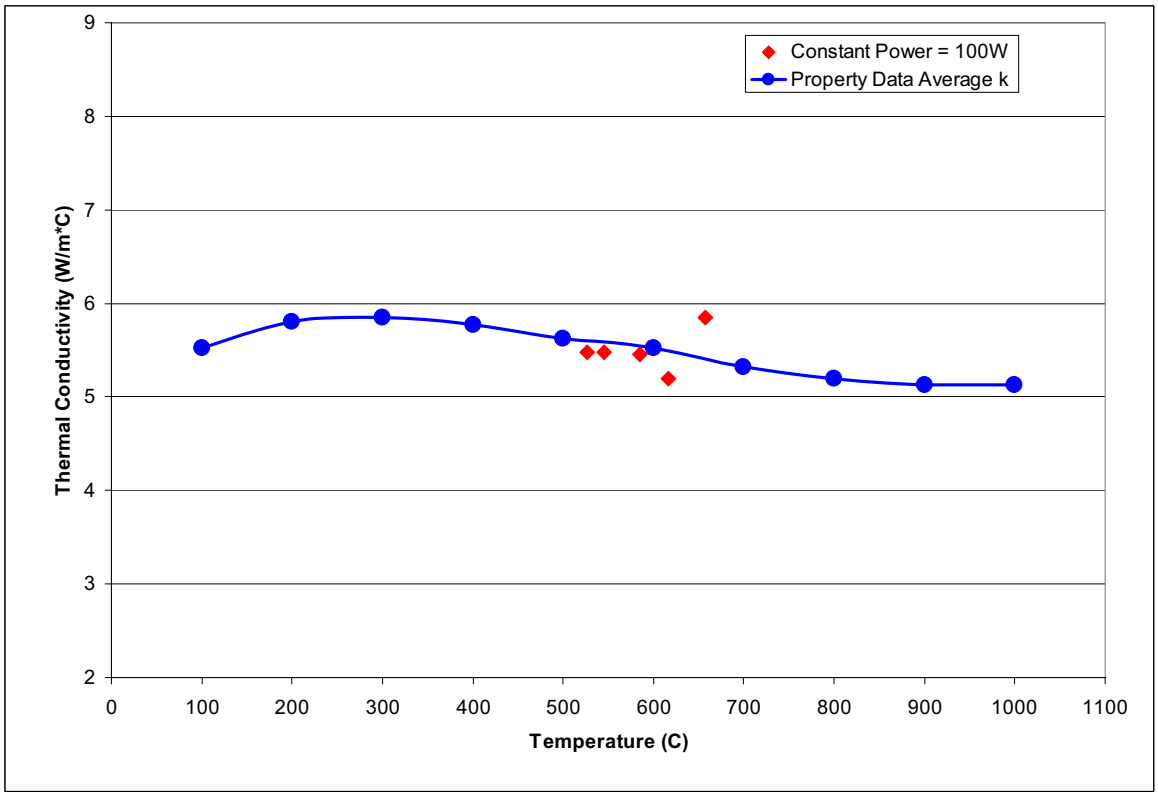

Figure 9. Measured CFOAM25 experimental thermal conductivity data from two-thermocouple approach compared with average values obtained with laboratory systems

2.1 Discussion

With continued validation testing still underway at INL's HTTL, it was found from early results, that thermal conductivity can accurately be measured from the proposed two-thermocouple method for steady state conditions. Several factors proved important for measurement accuracy, such as surface boundary conditions, supplied power, ΔT, and ambient temperature. Results show that, in general, higher ΔT values yielded better results for larger temperatures ranges. Values shown in Figure 9 were obtained over a temperature range from 500 - 700 ºC with a constant supplied power of 100W. These values ranged from 2% - 8% of the values in the material properties curve shown in Figure 9, and were within the 14% uncertainty range of the thermal conductivity calculation from the properties discussed in subsections 1.2.1-1.2.3. The accuracy of the constant power 100W data shown in Figure 9 is also within the possible experiment uncertainty, as discussed in the next section. Testing with temperatures above 700 ºC has revealed measurement limitations using the proposed two-thermocouple method and investigations to improve the test temperature range are still ongoing.

2.2 Uncertainty Analysis of Experimental Method

The uncertainty of the experimental method [7] is estimated for only the experimental measurements and the approach and setup uncertainties are not include within this analysis. Equation 8 is rearranged to evaluate the uncertainty impact of each measurement parameter shown in equation 9:

$$k = \frac{I \cdot V \cdot r^2}{4 \cdot \Delta T \cdot \pi \cdot r_o^{\,2} \cdot L}. \tag{9}$$

Where, $\dot{q}$ is defined as the product of measured current and measured voltage divided by the volume measurement, with I is the measured current, V is the measured voltage drop, r_o being the radius of the rod and L the length of the rod. Defining dk as the uncertainty of equation 8, the partial differentials can be taken of equation 9:

$$dk = \frac{V \cdot r^2}{4 \cdot \Delta T \cdot \pi \cdot r_o^{\,2} \cdot L} \cdot dI + \frac{I \cdot r^2}{4 \cdot \Delta T \cdot \pi \cdot r_o^{\,2} \cdot L} \cdot dV + \frac{2 \cdot I \cdot V \cdot r}{4 \cdot \Delta T \cdot \pi \cdot r_o^{\,2} \cdot L} \cdot dr - \frac{I \cdot V \cdot r^2}{4 \cdot (\Delta T)^2 \cdot \pi \cdot r_o^{\,2} \cdot L} \cdot d\Delta T - \frac{2 \cdot I \cdot V \cdot r^2}{4 \cdot \Delta T \cdot \pi \cdot r_o^{\,3} \cdot L} \cdot dr_o - \frac{I \cdot V \cdot r^2}{4 \cdot \Delta T \cdot \pi \cdot r_o^{\,2} \cdot L^2} \cdot dL. \tag{10}$$

Dividing by k:

$$\frac{dk}{k} = \frac{dV}{V} + \frac{dI}{I} + 2 \cdot \frac{dr}{r} - \frac{d\Delta T}{\Delta T} - 2\frac{dr_o}{r_o} - \frac{dL}{L}. \tag{11}$$

Thus, the general uncertainty of the two-thermocouple experimental measurement method is:

$$\varepsilon_k = \sqrt{(\varepsilon_V)^2 + (\varepsilon_I)^2 + 2 \cdot (\varepsilon_r)^2 + (\varepsilon_{\Delta T})^2 + 2 \cdot (\varepsilon_{r_o})^2 + (\varepsilon_L)^2}. \tag{12}$$

In the above equation, ε_V is the voltage measurement uncertainty provided by the power supply manufacturer, ε_I is the current measurement uncertainty based on calibration numbers, ε_r is the uncertainty from radial distance measurement based on thermocouple dimensions and machining tolerances, $\varepsilon_{\Delta T}$ is the uncertainty from the ΔT measurement given by the thermocouple manufacturer, and ε_{r_o} is the uncertainty from the radius measurement, and ε_L is the uncertainty from length measurement. Table III shows the percentage of contributing uncertainty from each of these sources.

Table III. Uncertainty analysis

Error Source	Error Percentage
ε_V	2.10
ε_I	0.10
ε_r	8.36
$\varepsilon_{\Delta T}$	0.75
ε_{r_o}	0.10
ε_L	9.30E-2
ε_k	12.03

Table III indicates that the largest calculated error is from the placement of the thermocouples within the sample and measuring the exact location. The assumption of reading 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; so in general, the larger the diameter thermocouple, the more error that will be introduced. However, the estimated measurement error of 12.03% falls in closely to the 14% difference between upper and lower bound and average values obtained with laboratory material property measurement systems. It was noted that while the thermocouple position was fixed in this experiment, measuring ΔT and $\dot{q}$ played significant roles in experimental measurement accuracy. Also noted, that other possible factors were not included in this uncertainty analysis, such as contact resistance between thermocouple and sample, and heat transfer surface boundary conditions.

3 CONCLUSIONS

Methods for in-pile detection of fuel rod thermal conductivity are being investigated using a surrogate rod material. As discussed in this paper, evaluations are first being performed to investigate a method which used two positioned thermocouples to measure the temperature of two points within the surrogate rod while Joule heating is applied to the rod for volumetric heat generation. Temperature-dependent thermophysical properties of elongation, specific heat, and thermal diffusivity, were measured using standard laboratory equipment to calculate thermal conductivity of the first surrogate rod material tested, CFOAM25. The test setup was modified to examine the sensitivity of results to several parameters, such as boundary condition changes, power supply variations, and temperature variations. Thermal conductivity was calculated under each condition to view the measurement sensitivity of method. Key results from this paper are:

1- Temperature-dependent CFOAM25 sample elongation was measured using four different samples. Figure 2 shows the calculated temperature-dependent densities. Maximum upper and lower densities varied by 0.07% and 0.07%, respectively, from the average calculated value for these samples.

2- CFOAM25 specific heat capacity was measured using three different samples and one sample crushed into powder form. Results shown in Figure 3 indicated that maximum upper and lower data values vary by 9.7% and 10.2%, respectively.

3- CFOAM25 thermal diffusivity was measured using three different sample thicknesses that were each tested two times. Results shown in Figure 4 indicate that the average diffusivity value varied from the maximum upper and lower values by percentages of 4.7 and 2.5, respectively.

4- The thermal conductivity of the CFOAM25 rod material was calculated, shown in Figure 9, using average values from the measured properties plotted in Figures 2 through 4. Although not shown in Figure 9, maximum upper and lower values for this estimated average thermal conductivity ranged between 8%-14% and 6%-12%, respectively.

5- The thermal conductivity of the CFOAM25 rod was measured experimentally using the USU/INL proposed two-thermocouple method. Values obtained for supplied power constant at 100W between 500 - 700 ºC were found to be within 2-8% from the values calculated using average values from standard material property measurement systems.

A comparison, shown in Figure 9, of the method calculation to that of the properties calculation shows that the method's results are within the range of the property calculations and have similar trends over specific temperature ranges. While improvements to the method are currently being implemented, the proposed method's early test results suggest the viability of using surrogate fuel rods to improve in-pile thermal conductivity measurement techniques.

3.1 Further Considerations

As mentioned, continued testing of CFOAM25 is still underway at INL's HTTL. Further CFOAM25 testing will focus on expanding the temperature test range. Because input power and ΔT showed a close correlation and understanding that a higher ΔT provides generally better accuracies, results below 500 ºC might prove difficult to obtain with high accuracies using the two-TC method. Also, because CFOAM25 was a porous material, testing is also being conducted to validate the proposed method using a solid metallic rod where extended testing can be conducted.

Also, while not included in this paper, the project will also look at a measurement technique using hot-wire methods. Both surrogate rod materials, CFOAM and the metallic rod, will be used to assess the hot-wire methods.

4 ACKNOWLEDGEMENTS

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4.1 Product Disclaimer

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

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