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HOTLAB Conference

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ABSTRACT

The thermal properties of irradiated nuclear fuels are extremely difficult to evaluate experimentally and thus have rarely been measured successfully, in spite of the vital role these properties play in fuel performance. A technique based on a commercially available 'hot disk' instrument is being developed to support thermal property investigations for plate-type nuclear fuels. Theoretical analysis was performed in order to evaluate the instruments response to a multi-layered test piece and to support calibration. In addition, a scanning thermal diffusivity microscope is currently under implementation that will permit point-to-point determination of irradiated nuclear fuels.

1. Introduction

1.1 Settings

Thermal properties of nuclear fuel, specifically thermal conductivity, play a vital role in fuel performance. However, evaluation of these properties is both challenging and very expensive to determine experimentally. Furthermore, fuel performance codes must assume extrapolated thermal property values or values calculated employing the Neumann-Kopp approximation, even though there is insufficient data for pure elements in some cases at elevated temperatures. Thermal diffusivity of nuclear fuels that can be used to determine safe operating temperatures and aid in process control and quality assurance.

One of the most common methods of determining thermal behaviour of fuels as a function of temperature and burn-up is to instrument a fuel pin in reactor with a thermocouple placed along the fuel centreline. This particular method of determining thermal properties is limited to a number of research reactors in the world capable of instrumented fuel pins, and is additionally very expensive to conduct. Another common method of thermal behaviour determination is to conduct property measurements on irradiated fuel pieces after removal from the reactor. Often this requires three different measurement methods: calorimetry for heat capacity, dilatometry for density, and laser flash for thermal diffusivity. Errors associated with each of these measurements are compounded in determination of thermal conductivity. Furthermore, determination of separate effects such as burn-up and fuel temperature requires multiple samples, increasing the cost and time required for analysis with each sample. This particular method also involves the loss of gaseous fission products in the matrix, so that integral behaviour of the fuel is lost.

Two techniques are being developed at the Idaho National Laboratory that will permit the measurement of multiple properties of irradiated plate-type nuclear fuels. The first technique is based on a commercially available 'hot disk' instrument while the second is based on an in-house developed scanning thermal diffusivity microscope. These two techniques are the subject of this talk, and development and advantages of each will be discussed.

2. Hot Disk Technique

2.1 Apparatus Description

The 'hot disk' instrument is commercially available through Setaram instrumentation and will offer determination of in-situ thermal property measurement of irradiated fuel after it has been removed in-reactor. By in-situ, the fuel is not removed from the cladding used in the reactor. Two hot disks have been purchased, one that will allow measurement of fuel thermal properties at beginning of life out of reactor, and a second to place in a hot cell that will allow measurement of the properties at end of life. The technique allows determination of thermal conductivity, thermal diffusivity, and heat capacity in a single measurement. The Advanced Test Reactor (ATR) in Idaho offers a unique advantage of irradiating a fuel plate approximately 1.14 m long in the centre flux trap. This allows determination of thermal properties in a single fuel plate with different burn-up levels and temperatures, depending on the measurement position. Thus, a thermal property map as a function of these parameters can be developed for a specific fuel. The data can be assumed to be "in-pile" in nature, since the measurement includes the influence of fission products in the matrix, fission gas bubble formation (porosity), micro-cracks, and Frenkel defects. Thermal property relations including the influence of burn-up and temperature are then determined including both separate and integral effects in a single fuel plate. The hot disk is capable of measuring thermal conductivity from 0 to 100 $W \bullet m^{-1} \bullet K^{-1}$ in seconds and temperatures from -50 to 200 ^{o}C.

The technique is based on a modified transient plane source technique, thus why only lower temperature ranges are capable of being measured. Generally, a one-sided, interfacial heat reflectance sensor will apply a constant heat source to the sample, with the known current providing a small amount of heat. This small amount of heat will result in a slight interfacial temperature rise, on the order of 2 K. The increase in temperature will result in voltage drop in the sensor. The thermo-physical properties of the sample, specifically thermal conductivity and effusivity, are inversely proportional to the rate of change in sensor voltage, e.g. the more insulating the material, the steeper the voltage rise. A photograph of the Hot Disk apparatus is provided in Figure 1.

Fig 1. Photograph of the Hot Disk apparatus.

2.2 Experiment Description

For the current case study, a monolithic U-Mo fuel foil is clad in AA6061. The fuel foil is approximately 250 μm thick, while each side of the AA6061 cladding is approximately 563 μm thick, for an overall plate thickness of 1.38 μm. Since thickness measurements are an essential parameter in determination of the thermal properties employing this technique, ultra-sonic scans of the fuel plates are taken both before and after irradiation, so that a "min-clad" can be determined and the thickness of the fuel being back-calculated. An example of a typical plate-type fuel with a monolithic fuel foil is provided in Figure 2.

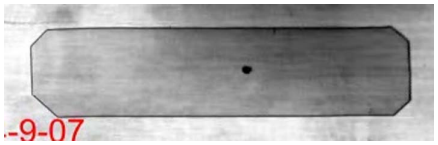

Fig 2. Example ultrasonic scan for a typical plate-type fuel with a U-10wt%Mo monolithic fuel foil.

2.3 Theoretical Considerations

The Green's function can be used to conveniently solve non-homogeneous problems in transient heat conduction in composite medium. First consider the following transient heat conduction problem in a multi-layer composite medium with internal heat generation, homogeneous boundary conditions at the outer surfaces, and perfect conductance at the interfaces, given by:

$$\alpha_i \frac{\partial^2 T_i(x,t)}{\partial x^2} + \frac{\alpha_i}{k_i} g_i(x,t) = \frac{\partial T_i(x,t)}{\partial t}, \quad in \quad x_i < x < x_{i+1}, \quad t < 0 \quad i = 1,2,\ldots M$$

,subject to the boundary conditions:

$$-k_1^* \frac{\partial T_1(x,t)}{\partial x} + h_1^* T_1 = 0, \quad at \quad x = x_1, \quad t > 0$$

$$\left\{ \begin{array}{l} T_i = T_{i+1} \\ k_i \dfrac{\partial T_i(x,t)}{\partial x} = k_{i+1} \dfrac{\partial T_{i+1}(x,t)}{\partial x} \end{array} \right\} \quad at \quad x = x_i, \quad t > 0, \quad i = 1,2,\ldots M-1$$

$$-k_M^* \frac{\partial T_M(x,t)}{\partial x} + h_{M+1}^* T_M = 0, \quad at \quad x = x_{M+1}, \quad t > 0$$

,and the initial conditions:

$$T_i(x,t) = F_i(x), \quad for \quad t = 0, \quad x_i < x < x_{i+1}, \quad i = 1,2,\ldots M$$

The appropriate eigenvalue problem for the solution of the above heat conduction problem is taken as:

$$\frac{d^2 \psi_{in}(x)}{dx^2} + \frac{\beta_n^2}{\alpha_i} \psi_{in}(x) = 0, \quad in \quad x_i < x < x_{i+1}, \quad i = 1,2,\ldots M$$

and subject to the boundary conditions:

$$-k_1^* \frac{d\psi_{1n}}{dx} + h_1^* \psi_{1n} = 0, \quad at \quad x = x_1$$

$$\left\{ \begin{array}{l} \psi_{in} = \psi_{i+1,n} \\ k_i \dfrac{d\psi_{in}}{dx} = k_{i+1} \dfrac{d\psi_{i+1,n}}{dx} \end{array} \right\} \quad at \quad x = x_i, \quad i = 1,2,\ldots M-1$$

$$-k_M^* \frac{d\psi_{Mn}}{dx} + h_{M+1}^* \psi_{Mn} = 0, \quad at \quad x = x_{M+1}$$

The solution to the multilayer transient heat conduction problem in terms of the composite medium Green's function $G_{ij}(x,t|x',\tau)$ is therefore:

$$T_i(x,t) = \sum_{j=1}^{M} \left\{ \int_{x_j}^{x_{j+1}} \left| G_{ij}(x,t|x',\tau) \right|_{\tau=0} F_j(x') dx' + \int_{\tau=0}^{t} d\tau \int_{x_j}^{x_{j+1}} \left| G_{ij}(x,t|x',\tau) \left[\frac{\alpha_j}{k_j} g_j(x',t) \right] dx' \right\}$$

$$in \quad x_i < x < x_{i+1}, \quad i = 1,2,\ldots M$$

where the composite medium Green's function is defined as

$$G_{ij}(x,t|x',\tau) = \sum_{n=1}^{\infty} e^{-\beta_n^2 (t-\tau)} \frac{1}{N_n} \frac{k_j}{\alpha_j} \psi_{in}(x) \psi_{jn}(x')$$

and the Norm is given by

$$N_n = \sum_{j=1}^{M} \frac{k_j}{\alpha_j} \int_{x_j}^{x_{j+1}} \psi_{jn}^2(x') dx'$$

where $\psi_{in}(x)$ and $\psi_{jn}(x)$ are the eigenfunctions and the βn values are the eigenvalues of the eigenfunction. The solution can be rewritten for a six layer system with heat generation only in the second layer and constant temperature initial condition (F=0), but the lengthy derivations are not discussed here.

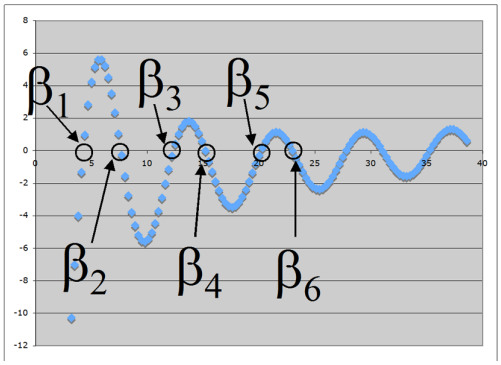

Fig 3. Example of the eigenfunction values for a six layer geometry.

Based on the geometric parameters measured via ultrasonic scanning and by assuming k/a values, the six eigenfunction values can be calculated, an example of which is provided in Figure 3. With these values, the temperature response can be calculated for the assumed k/a values. The analytical result is determined for several k/a values and compared with a measured response to evaluate which eigenvalues provide the best fit. A sensitivity study is currently in progress to determine the technique's resolution for the sample types of interest.

3. Scanning Thermal Diffusivity Microscope

3.1 Apparatus Description

The scanning thermal diffusivity microscope will yield a very unique capability for determining local changes in thermal diffusivity as a function of irradiation conditions. A photograph of the instrument showing pertinent features is provided in Figure 4. The instrument has been modified for remote application and is currently undergoing qualification and calibration studies before being installed into a hot cell in approximately 2-3 months. This is a laser based technique, and when fully functional, the technique will yield very a very detailed thermal diffusivity map of the irradiated material surface within resolution of 50 μm. Currently, the instrument can determine thermal diffusivity determination for a very wide range of materials, such as stainless steel on the low thermal diffusivity end to copper on the high diffusivity end, for temperatures up to 450 ^{o}C. These temperatures are adequate for research reactors, but higher temperature capabilities are being explored at the moment to represent other conditions, including power and fast reactors.

3.2 Experiment Description

Initial check-out and calibration tests have been performed on the instrument employing a Zr standard, with results closely matching those predicted by theory. Examples of these results are provided in Figure 5. Two Zr samples were measured with both the STDM and a laser flash thermal diffusivity (LFTD) method. The slope of the line passed through the data points as a function of temperature are very close to one.

4. Conclusions

Two techniques are being developed at the INL for thermal conductivity and diffusivity measurement of irradiated fuel. The Hot Disk technique will allow thermal conductivity, thermal diffusivity, and heat capacity determination of an irradiated fuel with both integrated and separate effect behaviour without instrumentation of the fuel. The Scanning Thermal Diffusivity Microscope technique will allow very detailed thermal diffusivity characterization of irradiated fuels up to 450 ^{o}C

5. Acknowledgement

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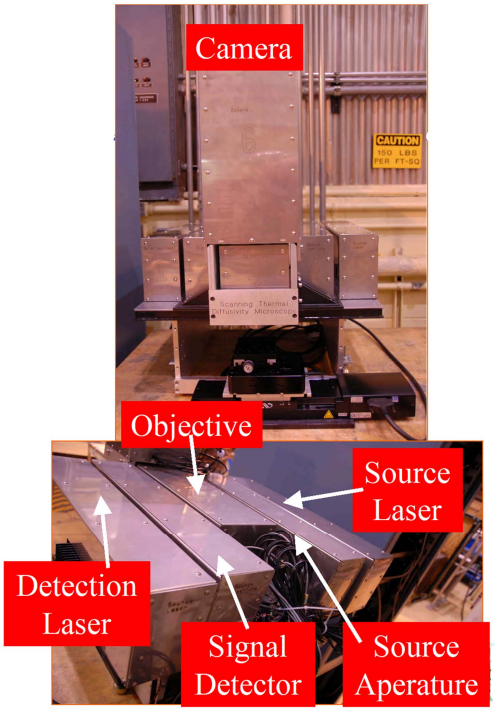

Fig 4. Photograph of STDM prior to installation in the hot-cell showing pertinent features of the device

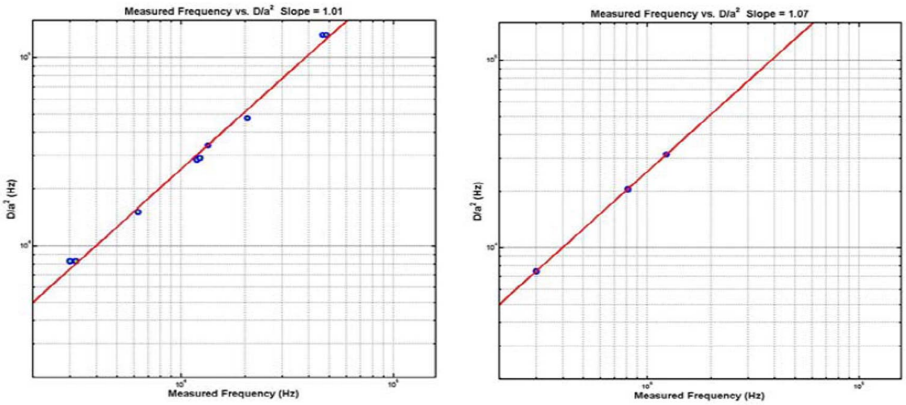

Fig 5. Initial calibration tests performed on a Zr standard showing good correlation between measurements made with the STDM and those made with LFTD