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Mesoscale Thermal Transport Measurements of Multi-phase and Porous Nuclear Fuels Using a Square-wave Pulse Thermoreflectance Technique

Scott Middlemas[1], Zilong Hua[1], Vinay Chauhan[2], W. Tanner Yorgason[3], Robert Schley[1], Cynthia Adkins[1], Tsvetoslav Pavlov[1], Marat Khafizov[2] and David Hurley[1]

[1]Idaho National Laboratory, Idaho Falls, Idaho, United States, [2]The Ohio State University, Columbus, Ohio, United States, [3]Arizona State University, Tempe, Arizona, United States

The safe and efficient operation of nuclear reactors require accurate knowledge of peak temperatures in the fuel assemblies. The temperature profiles are governed by the thermal transport properties of the fuel, namely the thermal conductivities (k) and thermal diffusivities (D). These properties can be very difficult to measure as they can vary considerably from the measured bulk values of the fresh fuel, and quickly degrade with increasing burnup [1-3]. Laser-based techniques have been effectively used for non-destructive and non-contact thermal transport measurements of a wide variety of materials, including nuclear materials [4-6] that would otherwise prove too hazardous or difficult to measure otherwise. In this study, a new thermoreflectance technique known as square-pulse transient thermoreflectance (SPTR) is described and used to determine the mesoscale thermal diffusivity of both uranium sesquisilicide (U_3Si_2) and uranium nitride (UN) phases in a composite fuel with micron level spatial resolution [7]. This technique employs a rapid train of square-wave pulses from an excitation laser to create a periodic heat flux on a gold coated sample surface. Surface adsorption results in transient film temperatures and hence rapid fluctuations in thermoreflectance that can be measured via a detection laser coupled with a digital oscilloscope, as shown in Fig. 1. The lasers are coaxially focused on the sample surface, allowing for a sample measurement area of a single convolved laser spot size (~2 μm).

A sensitivity analysis was conducted to identify key measurement parameters of this technique using reference materials with a range of thermal conductivities comparable to those of both ceramic, composite, and metal nuclear fuel types (1.4 – 27.2 W/m-K). The reference materials were measured using the new technique (see Fig. 2) as well as a spatial-domain thermoreflectance technique (SDTR) previously reported for a comparison [8]. Additionally, measurements of several U_3Si_2 and UN phase regions of a polished UN/U_3Si_2 (70/30 vol.%) sample were taken, and the resulting calculated D values are reported, with both techniques showing excellent agreement between samples. This technique was used to scan a multiphase region at 5 micron increments to generate a local diffusivity map, demonstrating the utility of the techniques for measuring thermal transport properties in specimens with precipitates and secondary phases. Furthermore, the technique is currently being applied to measure thermal properties of Fast Flux Test Reactor (FFTR) irradiated metal fuel specimens whose porous microstructure make it very difficult to measure using other techniques. Comparison of the local scale measurements are compared with the pre-irradiated fuel samples to show the degradation of thermal transport in fuels due to pores from fission gas bubbles and other irradiation induced defects.

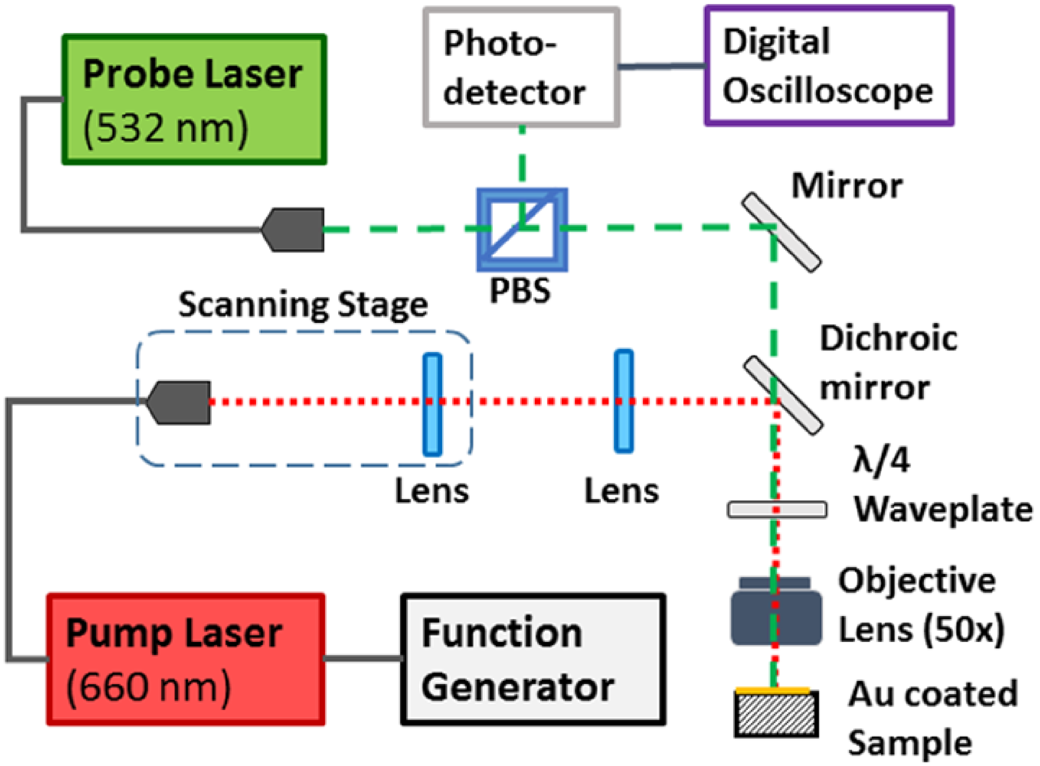

Figure 1. Experimental setup for square-pulse transient thermoreflectance (SPTR). A function generator supplies a square-wave pump signal to a 660 nm laser. A 532 nm probe laser detects changes in surface temperature via changes in thermoreflectance of the gold coated sample surface, which is relayed to a photodetector linked to a digital oscilloscope.

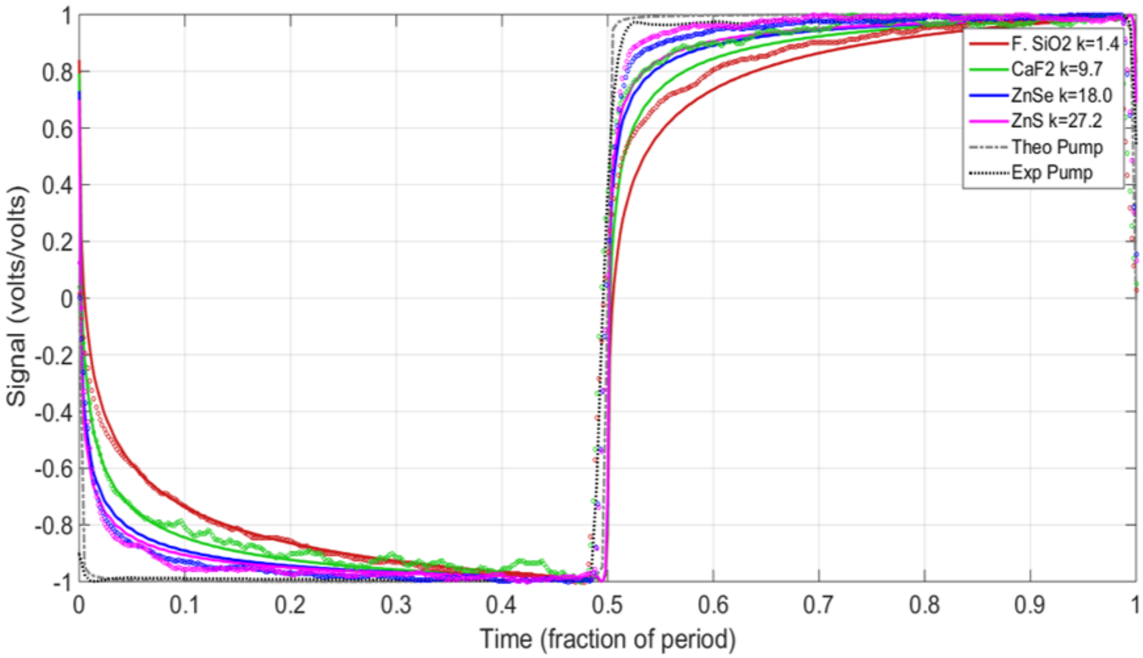

Figure 2. Experimental (markers) and theoretical (solid lines) thermal response waves excited by a pump laser at 10 kHz (black dotted line) for standard materials with a range of thermal conductivities, k (W/m-K). The excitation pump laser signal (Exp Pump) is shown as a reference (gray dash-dot line).

References

[1] Y.S. Kim, G.L. Hofman, S.L. Hayes, A.M. Yacout, Modeling of constituent redistribution in U-Pu-Zr metallic fuel, Journal of Nuclear Materials, 359 (2006) 17-28.
[2] V.V. Rondinella, T. Wiss, The high burn-up structure in nuclear fuel, Materials Today, 13 (2010) 24-32.
[3] M. Amaya, J. Nakamura, F. Nagase, T. Fuketa, Thermal conductivity evaluation of high burnup mixed-oxide (MOX) fuel pellet, Journal of Nuclear Materials, 414 (2011) 303-308.
[4] M. Khafizov, V. Chauhan, Y. Wang, F. Riyad, N. Hang, D.H. Hurley, Investigation of thermal transport in composites and ion beam irradiated materials for nuclear energy applications, Journal of Materials Research, 32 (2016) 204-216.
[5] J. Pakarinen, M. Khafizov, L. He, C. Wetteland, J. Gan, A.T. Nelson, D.H. Hurley, A. El-Azab, T.R. Allen, Microstructure changes and thermal conductivity reduction in UO_2 following 3.9MeV He^{2+} ion irradiation, Journal of Nuclear Materials, 454 (2014) 283-289.
[6] R. Cheaito, C.S. Gorham, A. Misra, K. Hattar, P.E. Hopkins, Thermal conductivity measurements via time-domain thermoreflectance for the characterization of radiation induced damage, Journal of Materials Research, 30 (2015) 1403-1412.
[7] S. Middlemas, Z. Hua, V. Chauhan, W.T. Yorgason, R. Schley, A. Khanolkar, M. Khafizov, D. Hurley, Determining local thermal transport in a composite uranium-nitride/silicide nuclear fuel using square-pulse transient thermoreflectance technique. Journal of Nuclear Materials, 528 (2020) 151842.
[8] D.H. Hurley, R.S. Schley, M. Khafizov, B.L. Wendt, Local measurement of thermal conductivity and diffusivity, Review of Scientific Instruments, 86 (2015) 123901.