Additive Manufacturing of Thermal Sensors for In-pile Thermal Conductivity Measurement

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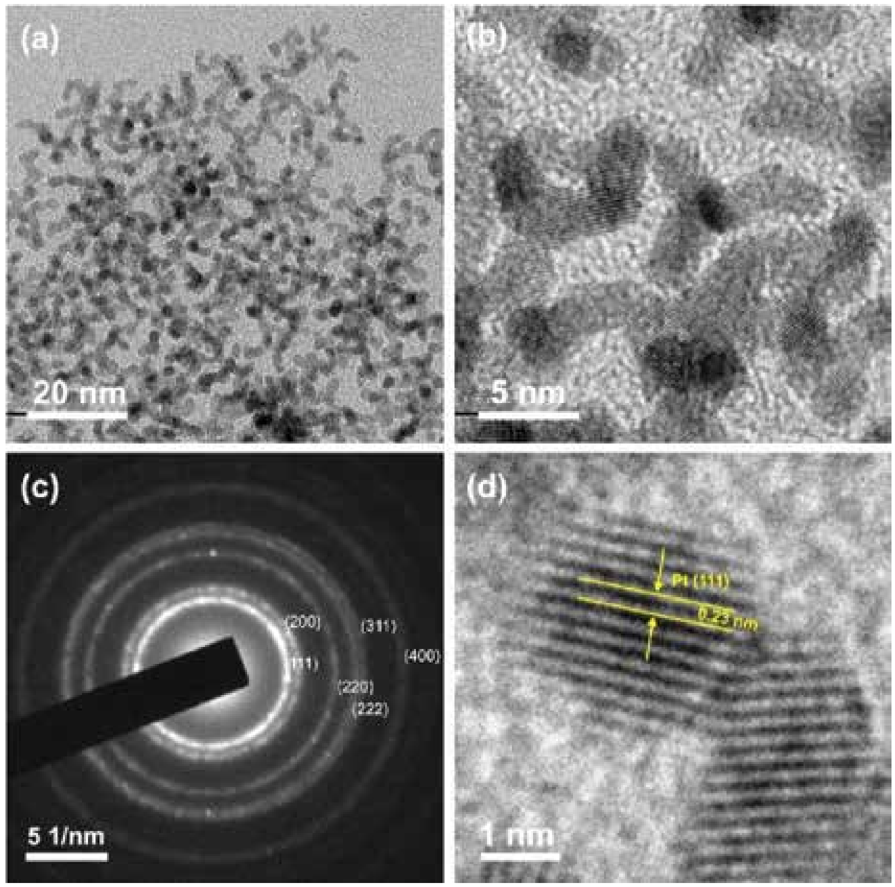

Figure 1. a, b)TEM and HRTEM d) images of the as-fabricated Pt nanoparticles. The corresponding SEAD pattern c) is also given.

The printed 3-omega sensors have great potential for improving the understanding of fuel thermal transport properties through in-pile thermal conductivity measurement.

Thermal conductivity is one of the most important fuel properties driving heat transfer performance as well as temperature distribution in nuclear fuel [1]. Thermal conductivity is determined by materials' physical structure, chemical composition, and thermodynamic state. These factors are strongly affected by a variety of physical processes in nuclear fuels, such as large temperature variations, species diffusion, neutron capture, and microstructure evolution [2,3]. Fuel thermal conductivity may also change substantially after removal from the reactor due to resulting changes in the material after the dynamic conditions of irradiation are removed. As a result, it becomes important to gain a more complete understanding of thermal transport as a function of time-temperature-burnup [4]. To do so requires accurate, spatially resolved

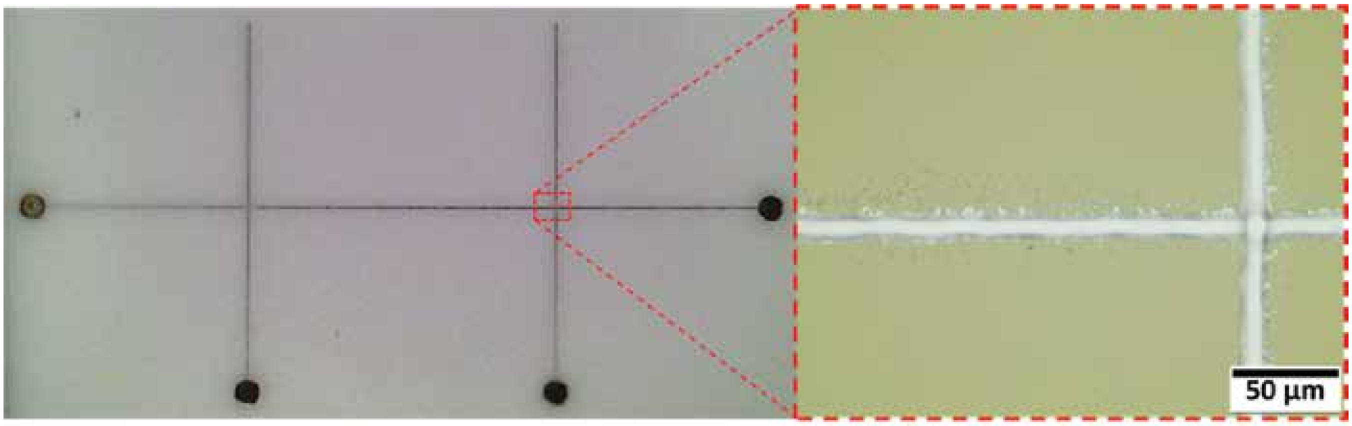

Figure 2. Microscope images of printed 3ω sensors.

in-pile thermal conductivity measurements. The majority of the fuel thermophysical properties measurements to date, including thermal conductivity, have been performed out of pile in a hot cell. In-pile measurement of thermal conductivity presents a significant challenge due to the space constraints, the difficulties to realize nonintrusive sensor implementation and the extreme environment of in-pile irradiation and temperature. Hence, very few thermal conductivity methods have been implemented in-pile, and these methods are often intrusive and fail to yield accurate fuel thermal conductivity.

Project Description

The goal of this project is to develop advanced 3ω sensors that can be tightly integrated with nuclear fuel system using advanced manufacturing methods to perform in pile thermal conductivity measurement. To determine the thermal conductivity, we will use the 3ω method.

A metal sensor directly printed onto substrates representative of nuclear fuel materials serves as both a heater and a temperature sensor. The heater is driven by AC current at frequency ω, which produces a localized alternating temperature change through periodic Joule heating at frequency 2ω, with tunable heat penetration depth controlled by the current frequency. The 2ω temperature change of the heater results in changes of its electrical resistance at frequency 2ω and a corresponding third harmonic component of the heater voltage (3ω voltage) which we can measure using a lock-in amplifier. This frequency-modulated thermal conductivity measurement offers real advantages over other methods for obtaining temperature-dependent thermal conductivity since we can confine the AC temperature field to the region of interest and minimize the influence of radiation heat loss.

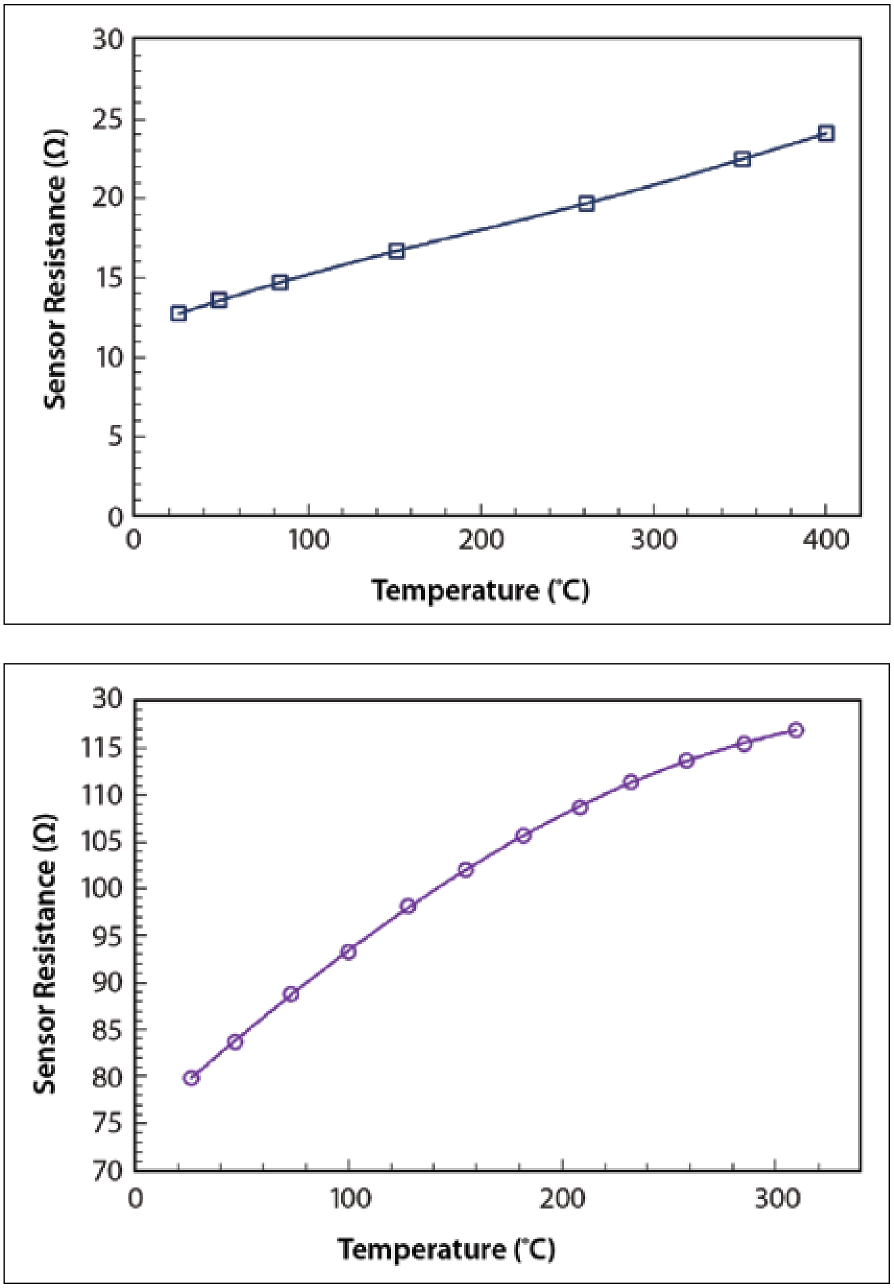

Figure 3. Resistance vs. temperature for aerosol jet printed silver (top) and platinum (bottom) 3ω sensors.

The project outcome will advance scientific knowledge of the in-pile performance for sensors fabricated using additive manufacturing. Furthermore, the insight gained will significantly accelerate the deployment of additive manufacturing to fabricate a broad range of sensors and instrumentation for both in-pile and out-of-pile measurements. The research outcome will have a broad impact on a number of DOE-NE initiatives including Fuel Cycle Research and Development, the Transient Reactor Test Facility, and Advanced Modeling and Simulation. This transformative sensor manufacturing process will advance sensor research and development activities in various areas of importance to DOE including research associated with the Advanced Test Reactor (ATR) programs, the Transient Reactor Test Facility (TREAT) restart, Light Water Reactor (LWR) programs, and spent nuclear fuel storage.

Accomplishments

Silver and platinum are two sensor materials studied for this project. While silver ink is commercially available, there exists no commercial platinum (Pt) ink that can meet this project requirement. We synthesized the Pt nanoparticles using a wet-chemical bottom up approach. Pt nanocrystal was prepared by dissolving platinum precursor in an oleylamine, Oleic acid and 1-octadecene mixture. Pt inks were then prepared by dissolving 0.3 mmol of the-above-fabricated Pt nanoparticles in 2 mmol chlorobenzene. Thanks to the narrow size distribution of Pt nanocrystals, the Pt ink can be well printed using ultrasonic atomization in the aerosol jet printer. Figure 1 shows Transmission Electron Microscopy (TEM), and a Selected Area Diffraction Pattern (SAED) of the

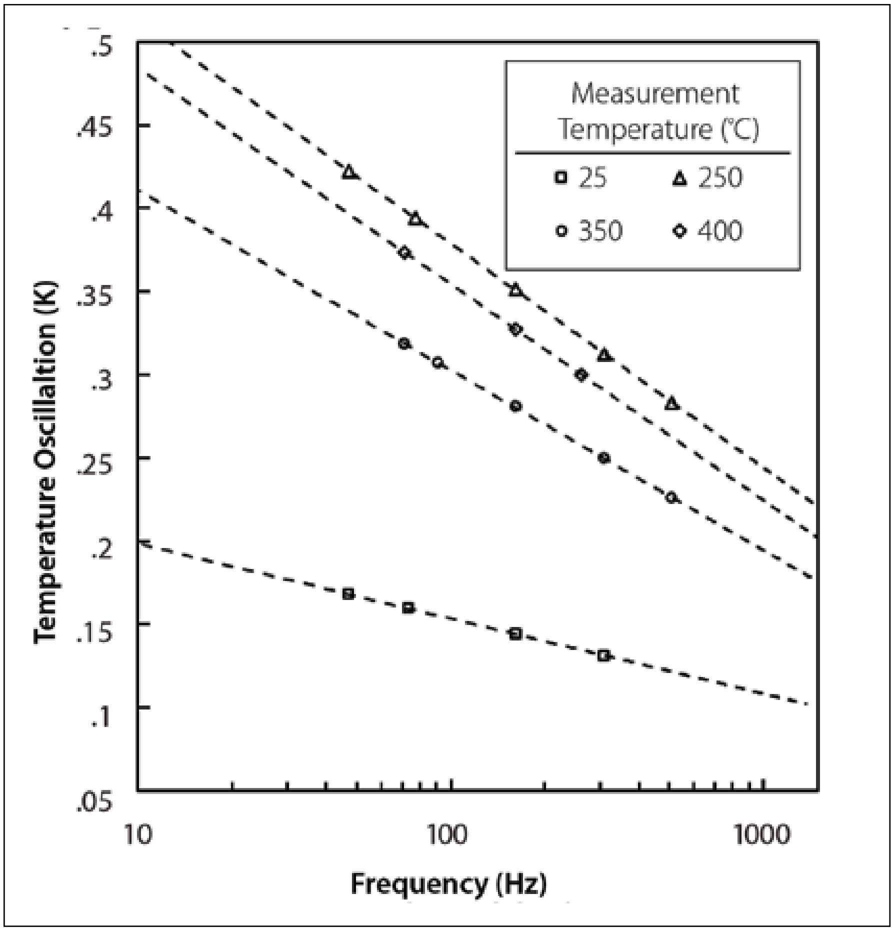

Figure 4. Temperature oscillation of the silver 3ω sensor on alumina vs. applied frequency for various temperatures.

platinum nanoparticles, confirming that Pt nanoparticles display good size and morphology uniformity with polyhedral structure.

In order to extract thermal conductivity with high accuracy, the width of the sensor must be much smaller than the thermal penetration depth. We optimized the aerosol jet printing parameter in order to print the 3-ω sensors with very fine microscale width. Figure 2 shows a printed silver sensor with 11.8 µm width with a small degree of overspray and non-uniformity in the sensor line width. Since the overspray consists primarily of <1 µm particles that are not in contact with the main sensor line, they do not influence the sensor electrical signal and contribute insignificantly to heat transfer. Besides optimizing printing parameters, another important task is to sinter the nanoparticles to obtain bulk-like electrical conductivity. In the case of silver, this was accomplished by sintering for at least 6 hours at a temperature at least 100°C greater than the target operation temperature. The primary material property governing the performance of a 3ω sensor is the temperature coefficient of resistance (TCR). Figure 3 shows electrical resistance as a function of temperature for a silver sensor and a platinum sensor. Room-temperature TCR values for

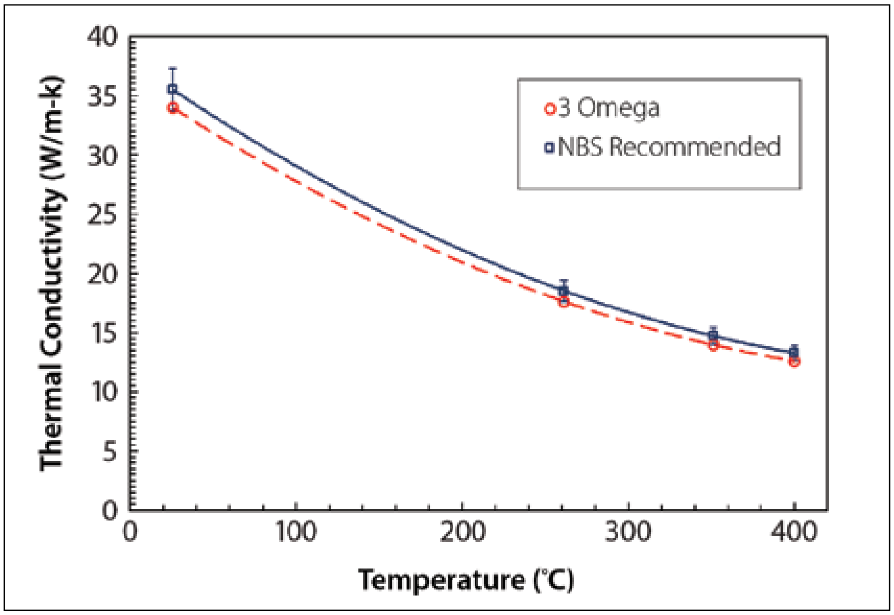

Figure 5. Thermal conductivity of alumina as measured by aerosol jet printed silver 3ω sensors and the recommended values reported by the National Bureau of Standards (NBS) [5].

aerosol jet printed silver sensors are 0.0025 K^{-1} on average. This is lower than that of bulk silver (0.0038 K^{-1}), which is to be expected of the printed sensors due to their nanostructure and size. Similarly, the TCR of the printed platinum sensors is ~0.0026 K^{-1}.

We have performed 3ω thermal conductivity measurements on alumina substrates, which have thermal conductivity values roughly in the same range of advanced nuclear fuels. Based on the alumina substrate thickness of 635 μm and the sensor width of 15 μm, the range of frequencies applicable to the 3ω slope method is ~60 to ~700 Hz. Figure 4 shows the temperature oscillation as a function of frequency for different temperature measurements. The results show an excellent linear dependence of temperature on the logarithm of applied frequency in the frequency range outlined above – a fundamental requirement of the 3ω slope method. The amplitude of temperature oscillations varies slightly for the different temperatures studied because slightly different current was used at each temperature. The thermal conductivity of the substrate can be determined based on the slope $dV_{3\omega}$ d(In9ω)) of the third harmonic voltage $V_{3\omega}$ to the natural logarithm of frequency ω using the following equation:

$$K = \frac{(TCR)\,V^{3}_{1\omega}}{4\pi l R}\,\frac{d(\ln(\omega))}{dV_{3\omega}},$$

where $V_{1\omega}$ is the voltage across the sensor at the fundamental harmonic, l is the sensor length, and R is the sensor resistance at the temperature of interest. Figure 5 shows the temperature-dependent thermal conductivity measurement results up to 400°C. The thermal conductivity of 99.5% pure alumina measured using the printed 3ω sensor agrees within about 5% of the value reported by the National Bureau of Standards. The excellent agreement validated the accuracy of the printed 3ω sensor, and demonstrates promises of the printed sensors for in-pile thermal conductivity measurement.

Future Activities

We have completed the first round of irradiation on the printed sensors at NC State PULSTAR Reactor. Our future work will focus on post irradiation examination on the sensors irradiated at NC State. We will further improve sensor high-temperature thermal stability in order to extend the thermal conductivity measurement to higher temperatures. We will prepare sensors for irradiation experiment at MIT Research Reactor with significantly increased neutron flux and dose.

Publications

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Idaho National Laboratory	Dave Hurley (co-principal investigator), Zilong Hua (collaborator)
University of Notre Dame	Nick Kempf (collaborator), Yanliang Zhang (principal investigator)