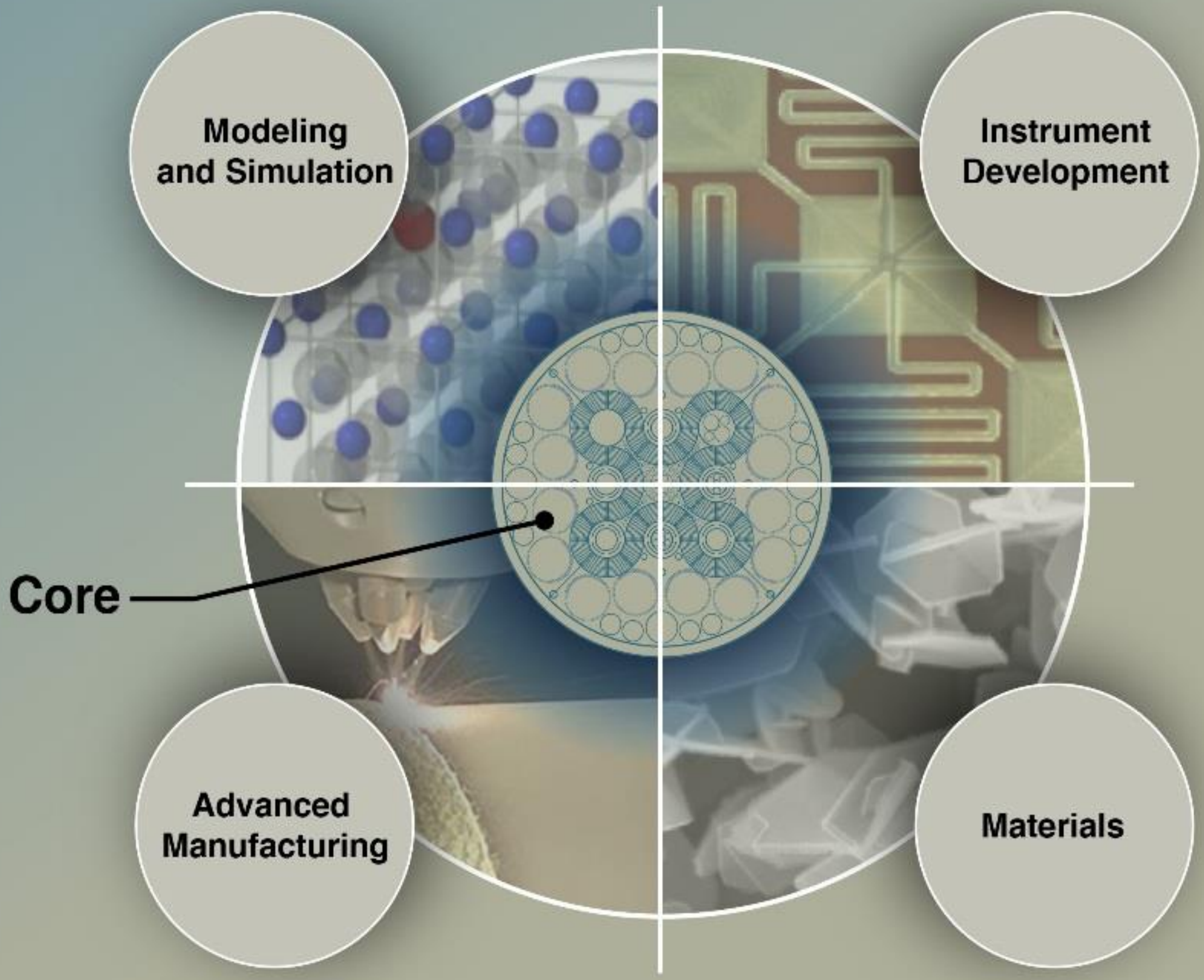
In-Pile Instrumentation Initiative



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SUMMARY

The In-Pile Instrumentation Initiative (I3) will conduct research needed to develop and deploy unique instruments to characterize the behavior of fuels and materials during irradiation tests conducted by Nuclear Energy (NE) programs within the Department of Energy.

(Vision) I3 will provide real-time, accurate, spatially resolved information regarding performance of fuels and materials that can be directly tied to microstructure.

This vision is reflected in the research direction of several NE programs that have identified a need to correlate material performance with evolving microstructure. Currently, these relationships can only be inferred through examination of materials in a post-irradiation environment. Realization of this vision will involve a sharp departure from the current approach that prioritizes instrument reliability over delivering optimal measurement capabilities.

To realize this vision will require technical breakthroughs in several key areas, including materials science, modeling and simulation, advanced manufacturing, and instrument development and deployment. Insights gained from this initiative will be critical for advancing a number of NE objectives. Examples include expediting the development of accident-tolerant fuels, providing a deep understanding of materials suitable for the extreme environments associated with new reactor technologies, and providing unparalleled validation metrics for new science-based fuel performance codes.

This Initiative is unique in that it will conduct research to address critical gaps in technologies related to measurement and characterization of fuels and materials during irradiation testing. It does so through a new paradigm of instrument development based upon strong interdisciplinary scientific collaboration. This collaborative effort will be divided into four science-based thrusts:

1. The **Materials Science Thrust** will use traditional and combinatorial high-throughput materials development to rapidly screen application-specific sensor materials. This will accelerate discovery, improve the resulting performance of the selected materials, and enhance the quality of data generated by in-pile sensors.
2. The **Advanced Manufacturing Thrust** will develop additive-, micro-, and nano-manufacturing techniques that will enable direct integration of both passive and active sensors onto fuels and materials.
3. The **Instrument Development and Deployment Thrust** serves as a focal point for combining the results of the other thrusts to transition prototype sensors into robust instruments that are ready for installation in the Nuclear Science User Facilities (NSUF) test reactors.
4. The **Advanced Modeling and Simulation Thrust** will develop tools and capabilities to predict the behavior of sensors and sensor materials in-pile. These capabilities will improve reliability, survivability, and availability of needed measurements during in-pile measurement campaigns.

The resulting in-pile instruments will provide essential information regarding evolution of the structure-property relationships, resulting in more direct measurement of key phenomena, fewer uncertainties, and greater access to real-time data. Critical outcomes of this Initiative will include the following:

- Disruptive capabilities for in-pile measurement of materials behavior
- Reduced development lifecycle for new in-reactor instrumentation
- Sensors that employ multiple methodologies (multimode) to improve data quality
- Smaller length-scale data that will provide insights into radiation-induced property evolution
- Access to in-pile material behavior that can’t be captured in a post-irradiation environment
- Connect changes in materials properties to changes in chemistry and microstructure.

Individually, these advances are important; collectively, they represent a step change in capability over current in-pile instruments. They are also responsive to the complex, in-pile measurement objectives identified by NE. By providing technologies that can be rapidly developed and customized to suit needs of individual irradiation tests, this initiative assists in addressing the needs for reactor and fuel cycle technologies identified in NE's Imperative 1, *Extend Life, Improve Performance, and Maintain Safety of the Current Fleet*. In addition, the Initiative will provide key and enabling capabilities to support the goals of Imperative 2, *Enable New Builds for Electricity Production and Improve the Affordability of Nuclear Power*. It will also provide a key element to the Gateway for Accelerated Innovation in Nuclear (GAIN) program by providing the nuclear energy industry with access to new monitoring technology and advanced validation of modeling and simulation.

The research direction of this Initiative addresses a coordinated set of needs that have been identified through engagement with stakeholders from the NE research and development (R&D) programs. The advanced sensors and instrumentation needs also integrate public-private partnership requirements into the overall set of R&D activities performed. The foundation of the Initiative is based on the four thrusts mentioned above. While each thrust addresses key technological questions, the overarching structure of the Initiative encourages strong engagement between thrusts. In addition to the four thrusts, there are five broadly defined R&D activities that have been identified by stakeholders from NE programs:

1. Measurement of field properties (neutron flux / pressure / temperature)
    - Accurate measurement of field properties is required for assessment of the effects of neutron damage, monitoring fission gas pressure buildup, and understanding the evolution of microstructure with temperature.
2. Measurement of materials properties
    - Thermal and mechanical properties of nuclear fuel and cladding materials play an important role in determining the operation characteristics of fuel assembles during normal operation and are crucial to understanding the response of fuel during accident conditions.
3. Chemistry and structure
    - Changes in chemistry over the lifetime of the fuel are closely tied to fission gas buildup, cladding embrittlement, and excessive corrosion, all of which lead to fuel failure.
    - Structure evolution, including fuel restructuring, crack formation, and swelling, has a profound influence on a range of material properties from thermal conductivity to mechanical strength.
4. Challenge activity - Power harvesting and wireless communication
    - Avoid complex design solutions necessary to interface electrical cables with the confinement barriers of nuclear systems. This activity by itself does not address a measurement need; rather, it will facilitate the other activities. Also, unlike the other activities, this activity is associated with only long-term goals.

By including key contributions from each thrust, the activities crosscut the thrusts and provide a very high level of integration within the Initiative. Additionally, an important and effective means of integration will come from project sharing and scientific visits by graduate students, post-doctoral fellows, and research leads from partner institutions.

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ACRONYMS

DOE	Department of Energy
NE	Nuclear Energy
I3	In-Pile Instrumentation Initiative
GAIN	Gateway for Accelerated Innovation in Nuclear
EPRI	Electric Power Research Institute
NEI	Nuclear Energy Institute
HTP	high-throughput
LENS	Laser Engineered Net Shaping

In-Pile Instrumentation Initiative: A Multidisciplinary Scientific Approach for Characterization of Fuels and Materials

1. INTRODUCTION

Researchers from the U.S. Department of Energy (DOE) Nuclear Energy (NE) programs require advances in in-pile instrumentation and characterization capabilities to better understand the performance of both nuclear fuels and materials in high-radiation environments for the development, deployment, and continued safe operation of nuclear energy systems and technologies. Material science of fuels and materials is fraught with complexity due to large thermal gradients, nuclear reactions, and the continuous production of defects. In oxide nuclear fuel, for example, thermal gradients cause cracking and are a driving force for chemical transport, nuclear fission changes stoichiometry, and point defect production leads to the growth of voids and dislocations. Microstructure elements such as Frenkel pairs, grain boundaries, and dislocations are interrelated and are tied directly to local temperature and strain fields.

This complexity is represented in Figure 1-1, which shows the fuel parameters that influence thermal transport. In this example, thermal conductivity is a time- and space-dependent property related to local microstructure as well as the time-dependent operational characteristics of the reactor. Currently, this relation can only be made based on model predictions.

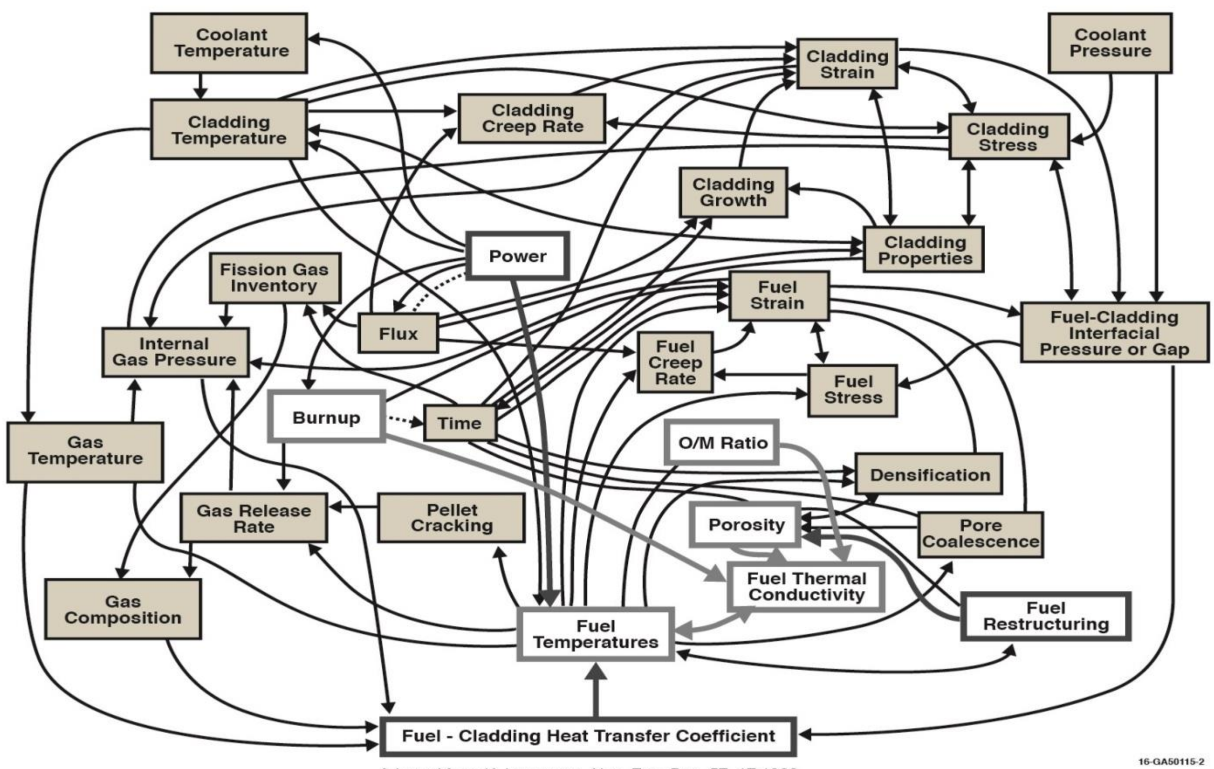

Figure 1-1. Complexity in nuclear fuel: parameters that influence thermal transport.

This complexity is further compounded by the recent observation that fuel thermal conductivity changes after removal from the reactor due to changes in defect density brought about by annealing and damage caused by self-irradiation.

The In-Pile Instrumentation Initiative (“Initiative” or “I3”) addresses these complexities associated with nuclear materials science by targeting dramatic advances in in-pile instrumentation that will enable the following:

- Direct access to the time-dependent, in-reactor environment (temperature, pressure, flux, etc.)
- Monitoring in-pile material behavior that cannot be captured in a post-irradiation environment.

For example, spatially resolved information on chemical composition could help modelers and fuel designers understand how fission gas is transported within a fuel assembly. Other examples include connecting the radial thermal conductivity profile to microstructure and connecting neutron flux to damage accumulation.

Improved in-pile characterization of material behavior must include extending the current characterization capabilities to allow for significant improvements in spatial resolution, data quality, and measurement accuracy. Achieving these goals will enable unprecedented access to the physical mechanisms that govern mesoscale materials science in extreme, in-pile environments.

This initiative will fundamentally change the nuclear materials development paradigm from “cook and look” to a systematic approach involving the development of new radiation-resistant materials and advanced manufacturing methods for fabricating small, multimodal sensors. These efforts will be directed by instrumentation deployment requirements and by multiscale and multiphysics simulations to address grand challenges associated with diagnostics of in-pile instrumentation. In addition to addressing the needs of NE, the proposed initiative squarely aligns with two nationwide research initiatives: The Materials Genome Initiative and The Advanced Manufacturing Partnership.

2. CONNECTION TO GATEWAY FOR ACCELERATED INNOVATION IN NUCLEAR (GAIN)

The Gateway for Accelerated Innovation in Nuclear (GAIN) initiative, as the organizing principle for the relevant DOE-NE programs, has a mission to provide the nuclear energy industry with access to technical, regulatory, and financial support necessary to move innovative nuclear energy technologies toward commercialization in an accelerated and cost-effective fashion. Recent technology-specific workshops hosted by GAIN, the Electric Power Research Institute (EPRI), and the Nuclear Energy Institute (NEI) suggested DOE laboratories develop monitoring technologies that could, for example, withstand very high temperatures, be used in opaque coolants, facilitate chemistry control, and increase the pedigree of modeling and simulation validation measurements (e.g. core condition monitoring). Development of technologies such as these through the In-Pile Instrumentation Initiative, either independent of or in collaboration with the advanced reactor design community, helps to address this need and thereby helps to fulfill GAIN’s mission.

The NE-Voucher program is managed by the GAIN initiative and, through a competitive process, offers the opportunity to have work performed on awardees' behalf at any of the DOE national laboratories or partner facilities. Funds are provided by appropriately aligned DOE-NE RD&D thrusts, and voucher recipients are required to provide a 20% cost share via funds-in or in-kind contributions. Voucher awardees who are focused on development of advanced instrumentation could be pointed to the Instrumentation Initiative for collaboration and performance of their proposed work. The GAIN initiative can facilitate alignment of industry needs with In-Pile Instrumentation Initiative capabilities to ensure development and commercialization of useful technologies.

3. CURRENT SITUATION

A number of NE programs will benefit from advanced in-pile instrumentation to explore material performance on mesoscale length scales. Information at these scales will provide the following benefits:

- Better inform new computation materials science models.
- Decrease uncertainty in fuel performance codes.
- Provide unique insight into mechanisms that control degradation of materials properties.

Today, sensor-development processes rely on expert knowledge and are both incomplete and, in many cases, represent significant risks for NE programs. The risks exist because relevant analysis and reactor testing is not performed to comprehensively qualify instruments prior to their use in harsh, in-pile environments. This results in early in-service failures of instruments, periodic loss of data from new instruments, and the continued reliance on existing instruments that offer reliability instead of desired measurement capabilities.

At the 2012 meeting of the American Nuclear Society, an experts group was queried on the state-of-the-art for in-pile characterization capability. The results are summarized in Table 3-1. Some incremental improvements in the Currently Available category have been achieved; however, it is striking that in the years following the meeting, none of the measurement techniques that were under development have transitioned into the Currently Available category.

Table 3-1. In-pile instrumentation capabilities that are either currently available or under development (unchanged since 2012).

Property / Field	***Methodology***	
Temperature	Meltwires / SiC Monitors/ Thermocouples	Currently Available at INL
Gas Pressure and Composition	Sampling	
Neutron Flux	Flux wires and foils	
Density/Dimensional	LVDT (Halden)	Currently Available Internationally
Elastic Constants	Loaded Creep/Tensile Specimen (Halden)	
Crack Growth	Direct Current Potential Drop (Halden)	
Gas Pressure	Pressure Gauge (Halden), Counter Pressure Gauge (CEA)	
Gas Composition	Acoustic Gas Monitor (CEA)	
Neutron Flux	Miniature Fission Chambers (CEA), SPND (Halden)	
Temperature	Ultrasonic Thermometer (INL), Thermoacoustic (INL), Fiber-Bragg (CEA)	Under Development
Thermal Conductivity	Needle Probe (INL)	
Density/Dimensional	Ultrasonic (INL), Thermoacoustic (INL)	
Neutron Flux	MicroPocket Fission Chambers (INL), Thermoacoustic (INL)	

It is clear that given the interest in fuel and cladding for advanced reactor concepts, and the renewed interest in accident-tolerant fuel, there is a great need for new sensors and analysis methods for in-pile characterization. Modern materials science methods, including those based on additive manufacturing, offer the potential to create a new generation of miniaturized sensors that could be widely distributed spatially in irradiation experiments and simultaneously measure a number of interdependent properties (multimodal measurement), which could be analyzed to elucidate the relevant irradiation conditions and the material response to those conditions.

4. APPROACH

New in-core instrumentation capability will require a paradigm-shifting methodology that connects sensor development and performance, material innovation, and modeling and simulation. Reaching this objective will involve combining advanced manufacturing with high-throughput combinatorial material science to develop radiation-resistant materials and create disruptive sensor designs. Additive manufacturing will facilitate the miniaturization of current in-core sensor technologies and will enable the development of new, miniaturized sensors capable of multimodal measurement. Combinatorial materials science will make possible the rapid development of robust sensor materials that can be used for more accurate measurements. The envisioned relation between additive manufacturing and combinatorial materials science is exemplified in several of the near-term R&D projects that are outlined in Section 6.

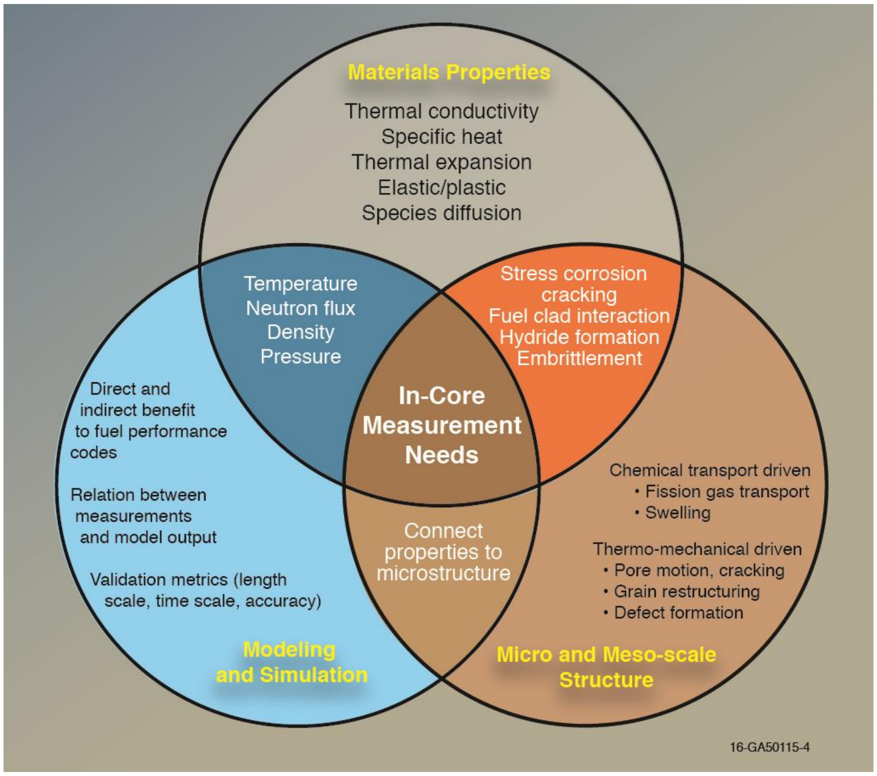

Figure 4-1. In-core measurement needs: the nexus between properties, modeling, and structure.

Ultimately, instrument development will be driven by the measurement needs that have been identified by NE programs. Figure 4-1 presents the measurement needs within the property-structure-modeling nexus for core materials. Measurements of temperature, pressure, and neutron flux can be performed with passive sensors and are perhaps the most straightforward measurements to make. Additionally, measurements of these field variables can be used to directly validate current modeling and simulation work. Measurement of materials properties will require active sensors and represent the next level of complication. The ultimate goal of this Initiative is to develop the ability to connect measured properties to chemistry and microstructure. This goal can only be accomplished by in-pile instrumentation enabled by advanced sensor technology.

Some of the needs outlined in Figure 4-1 can be met with only slight modification of current technologies. One illustrative example involves measuring the radial profile of the maximum temperature inside a fuel pellet using arrays of miniature melt wires. Other needs that connect microstructure to properties will require a transformational approach that directly connects instrument development and sensor performance. For instance, pore redistribution in an oxide fuel could be measured using an array of miniature ultrasonic transducers attached to the surface of the fuel pellet. However, several technical challenges must be addressed to make such a measurement possible. Example challenges include the development of radiation-resistant piezoelectric transducer materials, accounting for deleterious influence of chemical diffusion on transduction properties at high temperature, modeling the influence of porosity on ultrasonic velocity, and development of robust instruments that are ready for installation in NSUF test reactors.

5. SCIENCE-BASED THRUSTS

To address the technical challenges and logistical issues associated with a large research effort, the I3 is strategically organized around four science-based thrusts that are crosscut by five R&D activities. The motivation for the thrusts is to provide a structure that encapsulates major scientific disciplines that are central to achieving advances in measurement capabilities. Individual thrusts will combine scientists and engineers with complementary backgrounds to work on several R&D activities simultaneously. This structure will promote the flow of ideas, help identify roadblocks, and formulate required course corrections. This section provides a general discussion regarding the thrusts and outlines the Initiative's research strategy. A more detailed discussion concerning R&D activities is given in Section 6.

<u>The Materials Science Thrust</u> will be tasked with rapidly screening application-specific sensor materials for high temperature and radiation environments. This will accelerate discovery and improve the resulting quality of the selected materials. <u>The Advance Manufacturing Thrust</u> will develop additive-, micro-, and nano-manufacturing techniques that will permit direct integration of both passive and active sensor design into fuels and materials. <u>The Instrument Development and Deployment</u> Thrust will transition prototype sensors into robust instruments that are ready for installation in NSUF test reactors. <u>The Advanced Modeling and Simulation Thrust</u> will predict the in-pile behavior of sensors and sensor materials.

5.1 Materials Science

Materials science provides a foundation for the development of in-pile sensors and instrumentation. Development of advanced materials that can characterize the physical, chemical, and microstructural properties of fuels and materials and, at the same time, tolerate extreme operational environments (high temperature, high radiation flux, corrosive environment, etc.) is essential to the success of this Initiative. Understanding changes in transduction properties with temperature, radiation damage, and chemical diffusion will be of specific interest to this thrust. Sensor materials used to probe temperature, pressure, irradiation flux, microstructural evolution, thermal properties, and fuel chemistry will be considered. Both a near-term approach that utilizes existing materials and a longer-term approach that utilizes combinatorial materials synthesis will be needed. Near-term research will focus mainly on the effects of temperature and irradiation on sensing properties and strategies to mitigate degradation in sensor performance. Intermediate and longer-term research will target breakthroughs in materials for irradiation-tolerant sensing and packaging using a high-throughput (HTP) combinatorial methodology.

In the near term, I3 will emphasize pairing existing materials and sensor fabrication techniques with new sensor designs. Of primary concern will be understanding the influence of temperature and irradiation on sensor performance. Example issues to be addressed for high-temperature studies include chemical diffusion and thermal stresses between the sensor and the material of interest. Radiation experiments will be carried out initially using protons or heavy ions followed with neutron experiments. Because protons or heavy ions only damage a thin layer at the surface of the material, this will require developing fabrication and property characterization techniques appropriate for thin surface layers.

The intermediate and long-term strategy will employ a HTP combinatorial approach to create large numbers of compounds that can be tested in parallel. Because radiation testing is typically both time consuming and costly, the HTP methodology will enable rapid development and selection of candidate materials. This is particularly appealing for developing radiation-resistant materials. To accomplish this will require developing a novel combinatorial HTP workflow and tools appropriate for irradiation screening. As an example, a combinatorial HTP approach could be used to derive material combinations for new melt wires to further improve in-situ melt temperature resolution. This would provide an important test case that would demonstrate the advantages of a combinatorial HTP workflow.

The HTP process will utilize thin films sputter deposited from several independent sputter sources as well as additively printed materials. This approach is capable of producing both compositionally gradient samples and site-isolated spots. These samples can be characterized for composition (XRF), phase (XRD), and relevant transduction properties. The latter will be developed to enable case-by-case targeting of intrinsic sensing properties, and the first-pass data will be analyzed automatically, allowing for rapid screening of large compositional spaces to quickly identify regions of interest. These regions will then be explored in a finer compositional scale through subsequent deposition, computational modeling, and experimentation cycles.

The Materials Science Thrust is also central to relating instrument output to structure and chemical evolution. For example, it is easy to envision a piezoelectric sensor being used to measure changes in acoustic velocity; however, relating changes in acoustic velocity to changes in structure is not trivial because multiple mechanisms can influence results (e.g., porosity, texture evolution). Another example is an electrochemical sensor, where the electrical impedance between two electrodes can be used to monitor changes in temperature, chemistry, microstructure, etc. The challenge is developing a methodology that is (1) capable of separating these effects and (2) suitable for radiation damage studies. The role of materials science in this respect is to work with the other R&D thrusts to develop experiments that can relate the sensing information directly to microstructural as well as mechanical, thermal, and chemical properties.

5.2 Advanced Manufacturing

The mission of the Advanced Manufacturing Thrust is to develop optimal manufacturing methods and capabilities that enable fabrication of transformative sensor technologies for in-pile monitoring and in-situ analysis. The Advanced Manufacturing Thrust will advance state-of-the-art sensor manufacturing methods and leverage lessons learned from other industries, especially those related to sensor manufacturing for extreme conditions. The Advanced Manufacturing Thrust will provide tight and non-disruptive integration of sensors and fuel assembly, which is crucial to accurately monitor in-pile fuel behavior. A diverse set of advanced manufacturing methods and equipment will be established and adopted, enabling versatile sensor fabrication processes. This versatility will significantly enlarge sensor design space and accelerate sensor R&D life cycles. Additionally, advanced manufacturing techniques can facilitate fully integrated sensors of dissimilar materials onto or within a fuel assembly with minimized disruption that will enhance measurement accuracy. Advanced manufacturing can significantly accelerate the feedback loop of design, modeling, fabrication, and testing by reducing the traditionally time-consuming manufacturing and prototyping time frame.

An important area to be investigated under this thrust is using advanced manufacturing techniques to integrate multimodal sensors (i.e., resistive, piezoelectric, thermoelectric) into a single instrument. For instance, temperature sensors that employ multiple methodologies (i.e., Seebeck effect and IR thermometry) can be integrated into a compact sensor module. This is critical to obtain accurate in-pile information because multiphysics phenomena can significantly influence measurement accuracy of a single mode sensor. Additionally, advanced manufacturing techniques will provide a diverse set of tools to develop and deploy an array of sensors that will enable significant improvements in spatial resolution, data quality, and measurement accuracy.

The Advanced Manufacturing Thrust will draw input from the Materials Science Thrust and develop suitable manufacturing methods based on sensor material compositional, structural, and dimensional needs. For instance, a standard micro-fabrication process may be employed for thin films and used for electrodes and electrical isolation. Additive manufacturing methods may be ideal, if the materials produced are in particle form, to realize direct conversion and integration of sensor materials into devices of significantly enhanced functionality and complexity. Conventional brazing, welding, or other advanced joining techniques will be adopted if the sensor materials are in bulk forms.

The processing-structure-properties relationship in the manufacturing process will be revealed using multi-scale experimental characterization and theoretical modeling. It is crucial to develop fundamental understanding and effective control of the microstructure in order to obtain the desired sensor performance under in-pile conditions. For instance, piezoelectric materials require certain crystal orientations to deliver maximum efficiency, and thus the selected manufacturing process needs to produce or preserve the desired crystal orientation. Also, various sintering methods will be explored to control the microstructure, such as grain size, texture, and second-phase distribution. Comprehensive characterization will be performed to study the contacts/interface properties of dissimilar materials as a result of different manufacturing methods.

The Advanced Manufacturing Thrust can also assist materials development by providing alternatives to producing advanced materials of gradient compositions, nanocomposites, etc. For instance, a direct ink-writing combinatorial printer can produce binary, ternary or quaternary systems for high-throughput materials screening. Advanced materials processing methods such as laser sintering can fabricate materials of unique microstructures that will provide opportunities to tune and control microstructure and properties.

5.3 Instrument Development and Deployment

The mission of the Instrument Development and Deployment Thrust is to enable the use of innovative instrument concepts in irradiation test facilities by validating their expected performance. Validation is implemented through definition of test requirements, design of a testing device to evaluate performance, development of a test plan, and execution of irradiation testing and post irradiation examination (PIE) activities. This process will be applied to isolated sensors to aid in materials development and complete instruments to demonstrate performance characteristics.

The design and deployment of an instrument starts with the identification of the fundamental responses of the sensing element that require validation in an integrated test environment (e.g., for measurements of prototypical neutron flux/fluence, temperature, pressure, etc.). This leads, in turn, to the definition of test requirements, the need to identify suitable test facilities for different stages of instrumentation design and testing, and the start of the design cycle for one or more testing devices. The proposed sensor material or instrument will be integrated in a test device comprised of structural elements, cables, insulators, connectors and feedthroughs, signal processing electronics, and data acquisition systems. The response of the test device is then characterized using a combination of modeling and simulation, single effect tests (for example, heat cycling), and engineering analysis. Based on the results, an irradiation plan is developed, which may involve one or more of the irradiation test facilities within the NSUF. In some cases, inexpensive ion irradiation may be sufficient. Examples include studies involving the initial stages of materials development or tests aimed at understanding materials compatibility. Analysis and design are performed to satisfy safety and interface requirements in coordination with facility designers and operators. Necessary control qualification and validation processes are carried out, including preliminary testing, monitoring of the test device assembly, and pre-test verification of electronic components, signal conditioning, and data acquisition. The sensor is then tested according to the irradiation plan, and available real-time data is monitored and analyzed. Feedback is provided to the facility operators to optimize test conditions. Characterization of microstructure changes brought about by irradiation damage will be performed in the appropriate facility following irradiation tests. After test completion, sensor/instrument performance will be studied and compared with pre-test benchmarks.

The Instrument Development and Deployment thrust's focus is the final, crucial step of taking promising sensor technologies and proving them in the in-pile operating environment. This focus will harvest the materials and sensors developed through the Advanced Manufacturing and Materials Science thrusts and transform them into implementable in-pile instruments and materials.

5.4 Modeling and Simulation

The Modeling and Simulation Thrust will use state-of-the-art computational techniques to enable faster, more cost-effective development of sensor materials, manufacturing processes, and devices. Although modeling and simulation have found wide application in many areas of materials development and engineering design, their use in the area of in-pile instrumentation has been limited due to the unique challenges of the in-pile environment. The Modeling and Simulation Thrust will address this gap by using state-of-the-art computational techniques to understand how factors in the in-pile environment, such as high temperature and radiation damage, influence material behavior and device performance. This task will be conducted using computational techniques spanning length scales from the atomistic (i.e., Density Functional Theory, molecular dynamics) to the mesoscale (i.e., phase-field, diffusion modeling) to the engineering scale (i.e., continuum mechanics, heat equation). Atomistic length scale modeling will be carried out using existing commercial and open-source software tools, such as VASP, HOOMD-Blue, and LAMMPS. These will be augmented by new capabilities developed as part of the Modeling and Simulation Thrust, such as improved automated screening of relevant material properties for sensor design, and force fields and simulation protocols for predicting microstructure in advanced manufacturing processes. Mesoscale and engineering-scale modeling will be carried out using the Multiphysics Object Oriented Simulation Environment (MOOSE) and its suite of associated computational tools under continuous development by DOE, such as MARMOT and BISON.

Thee modeling and simulation thrust is closely connected to the Materials Science Thrust. For instance, several computational techniques are available to screen hundreds or even thousands of candidate materials with optimum properties and desired performance for different applications. These techniques can be used to help narrow the range of compositions and processing strategies explored in combinatorial materials science. In the short term, commercial computational thermodynamics tools such as Thermo-Calc Software can be used to select materials for sensor applications based on bulk thermodynamic properties. This will demonstrate how to use computational techniques in screening materials on the basis of key material properties, for example, melting temperature as a function of composition for temperature sensing applications.

Computational quantum mechanical simulation techniques, such as first-principles density functional theory (DFT), are common, cost-efficient, quick, and comprehensive methods to calculate material properties based on its atomic structure. Using DFT methods, an automated screening process will be developed to screen candidate materials for acoustic sensor applications based on piezoelectric coefficients. A similar DFT-based screening process will be used to screen thermoelectric materials for power harvesting applications based on Seebeck coefficient, electrical, and thermal conductivities.

Data from DFT calculations can also be used in conjunction with mesoscale simulation techniques such as the phase-field method, which simulates the evolution of materials at the microstructural level. These techniques can be used together to evaluate the effects of extended high-temperature operation on material properties. DFT will determine structural stability, estimated kinetic pathway, activation energy, and diffusion coefficients. The kinetic parameters from DFT will be imported to the phase field models to study diffusion processes on the micro and macro scales. Once developed, this methodology can also be applied to understand the effects of chemical diffusion on materials that may be used to develop thermal conductivity sensors in nuclear fuels and materials.

Modeling techniques such as molecular dynamics (MD), Monte Carlo, and rate theory have been applied to assess radiation effects on materials, but thus far have primarily been applied to structural properties. At the same time, there is a fundamental lack of scientific understanding of why certain sensor materials have high-radiation resistance. For example, the ceramic AlN is known to have a piezoelectric coefficient that shows high-radiation resistance compared with similar ceramics, but the reasons are unknown. Thus, a major effort of the Modeling and Simulation Thrust will be to apply computational tools to investigate radiation effects on sensor materials and properties. Key material properties of interest will include electrical resistivity (for thermal conductivity measurements), piezoelectric coefficients (for acoustic velocity and dimensional change measurements), and Seebeck coefficients (for power harvesting applications).

Modeling and Simulation is also strongly tied to The Advanced Manufacturing Thrust. In order to enhance advanced manufacturing capabilities, the Modeling and Simulation Thrust will investigate how to control the microstructure of materials produced using techniques such as inkjet printing. To optimize inkjet printing processes, MD simulations will be used to understand how the aggregation of nanoparticle inks dispersed in solution depends on the ink chemistry, particle shape and size, and solution choice. Simulations accelerated with graphics processing units (GPUs) will permit the screening of thousands of ink combinations, and the resulting morphologies will be characterized for cluster size distribution, porosity, and connectivity. MD simulations will also be used to evaluate the effects of post-printing sintering on the microstructures of inkjet-printed materials. The focus of these atomistic simulations is to provide fundamental understanding of the final microstructural features of a sintered ink and the resulting material properties. These simulations will be used to evaluate properties such as the grain size and orientation relationships in piezoelectric materials for ultrasonic sensors.

6. INSTRUMENT R&D ACTIVITIES

The reactor environment imposes many constraints (e.g., extremes of temperature and pressure, radiation effects, feedthroughs, geometries). The range of instruments to be considered for use in monitoring within the pile should include considerations not only for long-term, high-dose exposures—as would be experienced in steady-state irradiations in high-performance material test reactors such as the Advanced Test Reactor (ATR)—but also transient-testing environments provided by reactors such as the Transient Reactor Test Facility (TREAT). In transient-testing conditions, neutron damage is nearly negligible, though gamma heating can be quite significant and variable. Despite the many constraints, reactors and experiments can be highly configurable to address those constraints, provided early adoption of innovative measurement strategies is employed. Also, and unique to transient-testing irradiations, time scales of interest range from milliseconds to minutes, further dictating a departure from a one-size-fits-all data-collection strategy.

Each R&D activity is broken down into focus areas. These focus areas run the spectrum from easy-to-implement, incremental advances of existing sensors to medium- and long-term approaches that offer step changes in accuracy, spatial resolution, and radiation tolerance.

6.1 Field Properties

In the context of this program we refer to the measurement of field properties as the characterization of scalar (e.g., temperature), vector fields (neutron flux) and tensor fields (e.g., strain) that exist in the core of nuclear reactors and impact the performance of nuclear fuels and materials during in-pile irradiation tests. Field properties can also be considered as the 'operating conditions' of the irradiation test. This is complementary to the measurement of the materials properties, which are related to the characterization of the response of the fuels and materials to the above defined operating conditions.

A practical example discussed in depth in this section is the temperature field, which is the operating condition established as a result of heat generation and thermal transport. The other field properties considered relevant to this program are the neutron field, the pressure field and the strain field. All these properties are both space and time dependent, as in most cases they present a three dimensional distribution inside the reactor core and vary not only as a function of reactor power but also as a function of changes in fuel burn-up, coolant chemistry, corrosion and inelastic deformation (such as creep). When discussing instrumentation specification it is therefore fundamental to consider spatial resolution (which is in general directly related to sensor size, or invasiveness with respect to the measured properties) and sensor time resolution.

The field properties above have been identified as the most impactful based on the objectives of this research program and the needs of NE (R&D) programs. Most material properties are temperature dependent; hence their characterization requires the accurate definition of the temperature field. In addition, material performance during irradiation testing depends greatly on the damage caused by neutron capture events. Accurate knowledge of the local, time-dependent neutron flux intensity and energy spectra is therefore essential. The measurement of pressure in the fuel plenum can be tied to chemical transport of fission gas from the fuel interior to the fuel/cladding gap. Similarly, the measurement of strain and deformation is related to the characterization of mechanical properties. In both cases the development of sensors planned under field properties is an essential element for the design and implementation of more complex techniques addressed as part the measurement of material properties.

The activities presented below identifies key research tasks required to accelerate development and deployment of sensors with advanced capabilities in the near and mid-term (1-3 years) and enable rapid development of innovative methods and technologies in the longer term (3-5 years).

6.1.1 Neutron flux

Neutrons are responsible for many physical processes underlying the operation of nuclear reactors. The energy released from neutron interactions with nuclear fuels through fission generates the heat that is harvested to produce electricity in commercial nuclear reactors. There neutron interactions are also responsible for materials properties degradation that ultimately limit the lifetime of the fuels and materials that comprise a nuclear reactor. The accurate characterization of the neutron field is therefore essential for in-pile testing of these nuclear fuels and materials. This implies accurate spatial resolution (ie, local neutron flux parameters instead of core/zone averaged parameters) as well as time resolution (ie, online measurements instead of averaged fluence). Because of the dependence of most capture processes on neutron energy, characterization of both the spectra and energy-averaged flux intensity are essential. These challenging requirements define the need for a new generation of neutron sensors for in-pile tests.

In the cases where only overall neutron fluence should be known, passive methods are used to determine the neutron fluence after the sample has been removed from the reactor. Flux foils and wires fall under this category and are the subject of R&D Activity 1 outlined below. Also known as a neutron dosimeter, the flux wire or foil is simply a material of known composition and purity that is placed in a neutron field and later characterized and correlated to the integral neutron exposure. Implementation of advanced manufacturing techniques such as aerosol jet printing (AJP) will be used to initiate improvements in size, material selection and post-processing techniques for the next generation of neutron dosimeters.

Other DOE-NE programs have identified candidate new materials and fuels having performance evaluations that require real-time, high-accuracy, high-resolution, and spectrum selective neutron flux measurements. In R&D activates 2 and 3, focus will be on miniaturized flux detectors and solid-state neutron detectors. To realize improvements in terms of size, radiation tolerance, or resolution for these measurement systems, the research activities will necessitate interaction and coordination with other areas. Specifically, the design team will interface with researchers in the areas of materials science, modeling and simulation, and advanced manufacturing to ensure prototypes will function as intended and meet critical specifications without sacrificing performance.

6.1.1.1 R&D Activities

R&D Activity 1: Develop passive neutron dosimeters (flux foils) fabricated by advanced manufacturing.

The first short term activity considered is the development of dosimeters fabricated by advanced manufacturing, to be used when knowledge of the time averaged fluence satisfies the test requirements. Materials of different composition offer the potential of providing at least partial resolution in space (i.e., radial profiling) and spectrum (energy profiling).

Advanced manufacturing of neutron flux foils stands to maximize these capabilities thru miniaturization and flexibility in fabricating materials with mixed and accurately controlled composition. In addition, the potential to lower the fabrication cost and post-processing requirements offers a path to intensive deployment and commercialization. Short-term activities include the fabrication of an additively manufactured passive dosimeter prototype, neutronics modeling for design optimization, and the development of a specially designed read-out system. Testing in irradiation facilities (in particular TREAT and MITR) is being planned with a focus on the comparison of the performance with materials fabricated by standard processes that have been routinely used in material test reactors for neutron dosimetry.

R&D Activity 2: Real-time, miniaturized flux detectors development

Fission chambers and Self Powered Neutron Detectors (SPNDs) are commonly used methods for monitoring local neutron flux in real-time. The theory of operation is well understood, hence advancements will be pursed that leverage recent improvements in manufacturing capabilities. The use of advanced materials and fabrication techniques are expected to miniaturize flux detectors without sacrificing performance. This will lead to a larger array of real-time flux data for irradiation experiments. In addition, improvements in SPND design may be leveraged for dual use as a power harvesting device for wireless sensor operation. Self-Powered Gamma Detectors (SPGD) will also be explored in parallel research activities because of the similarities to SPND design and operation.

R&D Activity 3: Nano-structured Materials for Solid-State Neutron Detection

Compared to classical gas-filled or scintillating neutron detectors, devices using solid-state materials can offer advantages in terms of power efficiency, scalability, and response time. Since neutrons have very little interaction with many materials, converter layers with high neutron interaction cross-sections are used which produce detectable reaction byproducts that can be measured by solid-state materials. We will analyze both external and internal converter material configurations, with a focus on radiation- and heat-tolerant materials. Promising candidates include diamond, SiC, and carbon-based nanomaterials grown by plasma-enhanced chemical vapor deposition (PECVD). It may also be possible to use printable materials such as boron nitride (BN) and carbon nanotubes (CNTs). These are mid to long term activities (3-5 years) for which leveraging on the science based thrust is foreseen, in particular for the investigation of novel materials.

6.1.2 Temperature

Temperature is a key parameter of interest during fuel and material irradiations. The fuel and cladding properties that determine their performance are strong functions of temperature, as is the evolution of material microstructural. As such, measurement of temperature during irradiation is needed to characterize the performance of fuels and materials and to aid in interpretation of post-irradiation examinations. Depending on the type of test targeted, temperatures of interest could vary from nearly room temperature to over 2000 °C. The conditions in which the temperature measurement may be needed can also vary widely in terms of both radiation and chemical environment (i.e. water, liquid metal, high temperature steam, etc.). The specific test targeted is also important in defining temperature instruments specifications in terms of spatial accuracy and time resolution. As an example, deployment in a TREAT test would require fast response in order to follow the transient reactor power.

Melt wires and SiC temperature monitors to record peak temperature for passive tests and commercially available thermocouples (e.g., Types K, N, and C) to measure temperature have been available for decades but have significant limitations in terms of temperature range, radiation tolerance, reliability and accuracy. The R&D activities listed below will leverage current advancements in the areas of materials science, modeling and simulation, and advanced manufacturing to develop the next generation of temperature sensors for irradiation tests.

6.1.2.1 R&D Activities

R&D Activity 1: Develop advanced passive temperature monitors.

Passive temperature monitors are deployed in many irradiation capsules as an inexpensive method to collect information about peak test temperatures. The monitors are interrogated during Post Irradiation Examination (PIE) activities to determine the peak temperature received during the irradiation test. One method, melt wires, is based on the melting temperature of bulk materials. These melt wire assemblies are fabricated from pure metal powders or wires and provide little or no spatial resolution, in addition to measuring peak irradiation temperature only (no time history). A second method, SiC temperature monitors, is based on the electrical properties of SiC in a neutron field. The process to interrogate these SiC monitors during PIE requires a time consuming thermal cycling. The two innovative technologies considered in this program for the development of a new generation of passive temperature monitors are aimed at overcoming the current limitations.

The first activity relies on the use of advanced manufacturing techniques to print melt wire arrays that can provide spatially resolved information of the peak irradiation temperature when analyzed during PIE. This activity, will use advanced manufacturing techniques to improve the consistency of the temperature resolution while also miniaturizing the sensor and developing electrical readout methods for consistent and rapid PIE. A library of nuclear grade inks and compositions for depositing melt wires of varying composition and melt temperatures will be developed. It is expected this research, combined with neutron dosimetry research activities will lead to a multimodal, passive drop in temperature and neutron flux sensor.

The second activity is the use of SiC temperature monitors and an automated analysis process during PIE that would substantially reduce the time needed to interrogate the monitors. In addition, new geometries would allow deployment in a variety of configurations used for material characterization positions (i.e., TEM discs), which would allow for improved spatial resolution and a wider use in irradiation tests.

R&D Activity 2: Develop advanced thermocouples for temperature measurement during nuclear fuel and materials irradiation tests.

The use of thermocouples is the most common and validated technique for the real-time measurement of temperature in an irradiation test. However, commercially available thermocouples are limited to about 1050^{o}C for drift-free performance under neutron irradiation and have typical response times ranging from a few seconds to minutes. DOE has sponsored the development of the High Temperature Irradiation Resistant Thermocouple (HTIR-TC) to fill this technology gap. HTIRs in various design stages have been tested out of pile and deployed in MTRs, including fuel development experiments at ATR. The reference materials for the HTIR components are part of a patented design, based on Molybdenum and Niobium electrode pair and a Niobium alloy sheet. However, advances in material research have made available new doped alloys that offer enhanced performance for electrodes and sheets. The initial results suggested a substitute for the reference insulation material and as a result, R&D activities are still needed in order to optimize the materials selection in combination with the fabrication and heat treatment processes. While the optimization of HTIR is the focus of near term activities, a new approach that combines testing with combinatorial material science and modeling could lead to the discovery of additional advanced materials offering improved performance in terms of radiation resistance and mechanical response of the sensor assembly. The screening of candidate materials and the testing of identified candidates will be the focus of mid-term activities for advanced thermocouples development with the goal of providing a path to commercialization based on currently available fabrication processes. Application of advanced manufacturing techniques to the fabrication of thermocouples is considered as a longer term project with the ultimate goal of printing insulated junctions directly on nuclear fuel elements.

A second area of interest is the development of fast response thermocouples that can detect transient events during irradiation, such as those foreseen during transient operation at the TREAT reactor. The development of exposed thermopile junctions attached to the fuel cladding represents a faster solution to fuel research programs needs for accurate real time temperature measurement. The activities are focused on the selection of optimized material combinations for electrodes and substrate material and rely on the synergy between material science, modeling, high temperature testing and out of pile characterization (ie, metallurgical analysis).

R&D Activity 3: Develop solid state thermistors for temperature measurement during nuclear fuel and materials irradiation tests.

Proof of principle research activities conducted at the University of Wisconsin-Madison (UW-M) indicates that a specially designed, irradiation resistant, diamond thermistor may hold superior performance characteristics that could make it the temperature sensor of choice with respect to drift, response time and sensitivity. Currently the UW-M irradiation resistant diamond thermistor is limited to 800°C, but continued research could extend the upper temperature limit to 2000°C. In the near term, Activity 3 will conduct research on new approaches to connect electrical leads to the diamond-based sensor. In the longer term this research will evaluate diamond replacement materials to extend the temperature range. Prototype thermistors will be constructed and bench top tested. Prototypes that pass benchtop testing will be scheduled for irradiation testing.

A second approach in developing a solid-state, high temperature sensor is based on silicon carbide (SiC) p-n junction diode detector. Although more limited in terms of operation temperature, SiC diodes offer the potential of fast time response that could provide an adequate solution for transient test needs, such as those of the TREAT facility. The near-term research activities in this area will focus on feasibility assessment and proof of principle demonstration.

R&D Activity 4: Develop optical fiber based temperature sensors.

Fiber Bragg gratings (FBG) and long period grating fibers (LPGF) are currently being considered as potential multi-parameter miniaturized sensors for distributed temperature measurements at high temperatures. Novel materials for potential fiber gratings to be used in nuclear reactors are being investigated as part of enabling technologies within this program. This activity is a medium term project focused on development of a fiber optic temperature sensor following the successful demonstration of the applicability of the selected methods to in-pile measurement as part of enabling technologies (Section 6.4).

R&D Activity 5: Develop ultrasound temperature (UT) monitors.

Ultrasound based thermometry is an innovative technology that allows accurate distributed temperature measurement in-core. Previous activities have demonstrated the proof of principle of the method, including experimental validation obtained through the deployment in multiple irradiation tests. Despite the successes of the recently completed activities, research and development is still necessary to bring the UT sensors to the level of maturity that is required by the objectives of this program. In particular, the sensors would greatly benefit from the science based and multidisciplinary approach implemented in terms of combinatorial material science analysis, modeling and the application of advanced manufacturing processes for miniaturization.

Near term activities will focus on additive manufacturing of enhanced materials with complex geometries. Improvements to other sensor components, such as enhanced temperature sensitivity of waveguides and improved corrosion resistance of protective housings will be explored. In addition, improvement of UT specific electronics and signal processing techniques will be explored in coordination with performance testing both in and out of core.

6.1.3 Pressure

In-core measurement of pressures focus on two principal parameters: coolant pressure and fission gas pressure. Coolant pressure is of less interest as techniques exist for the reliable, real time measurement of this parameter and they are performed outside of the core. On the other hand, fission gas release is a key performance and safety parameter of interest during fuel irradiations and this program will focus on developing sensors that can measure pressure in the volumes surrounding materials and fuels being tested, normally in gas phase. There are several existing methods of measuring pressure that can be applied to in-pile tests, both during post-irradiation examination (PIE) and in situ.

Fiber optics and piezoelectric can be used to measure gas pressure, and these technologies have been selected as the focus for the development of innovative in-pile pressure sensors as part of enabling technologies (Section 6.4). The main measurement technique considered for this application is Fabry-Perot interferometry, where the deformation of a crystal (sapphire or quartz) due to external pressure is detected by monitoring shifts in the interference patterns of overlapping light waves. The detection of Rayleigh backscattering lights can also be used to measure the fiber deformation, and related it to the pressure acting on it. This method will be considered as second alternative since it is cross cutting with the development of radiation tolerant fibers (it’s used for status monitoring) as well as temperature measurement.

6.1.3.1 R&D Activities

R&D Activity 1: Develop a system to measure fission gas product total pressure using optical fibers.

This activity is a medium term project focused on development of a fiber optic gas pressure sensor following the successful demonstration of the applicability of the selected methods to in-pile measurement as part of enabling technologies (Section 6.4).

R&D Activity 2: Develop pressure sensors based on novel radiation resistance piezoelectric materials

This activity is a medium term project focused on development of piezolectric sensor following the successful demonstration of the applicability of the selected methods to in-pile measurement as part of enabling technologies (Section 6.4).

6.1.4 Strain and deformation

Dimensional changes occur in structures as a result of loads imposed by mechanical loads and the mechanical constraints to which they are subject. For nuclear fuel and materials the main loads are imposed by temperature gradients due to neutron and gamma heating, the pressure exercised by coolants and component weight. Dimensional changes result in deformation and/or strain, depending on the mechanical constraints and radiation induced swelling. There is a wide variety of technologies that are used in commercial and research application to measure deformation and strain. The first objective of the

R&D activities is to provide a comprehensive assessment and prioritization of the techniques applicable to in-pile measurements of nuclear fuel and materials. Those can be grouped in the following typologies:

- Electrical based sensors, where dimensional changes are related to variation in the impedance of an electrical circuit in the form of resistance, capacitance or induction depending on the design of the sensing element.
- Optical sensors, where light (including direct or reconstructed images) is used to characterize dimensional changes.
- Acoustic sensors, where changes in the transmission of sound waves are related to dimensional changes.

6.1.4.1 R&D Activities

R&D Activity 1: Development of LVDTs and capacitance sensors to measure deformation in-pile

The application of existing LVDT sensors to the measurement of fuel pin dimensional changes during irradiation will be considered for short to mid-term activities (1-3 years). This effort looks at both the diameter and elongation deviations of a fuel pin (e.g. swelling, crude build-up, thermal expansion, etc.) during irradiation experiments. One of the main challenges to be addressed is the development of a hermetic seal (i.e. piston cylinder) capable of withstanding the harsh environment of the reactor test channels – while translating in the axial direction. The long-term objective of this activity (3-5 years) is the development of miniaturized LVDT sensors thru the application of advanced manufacturing techniques.

In the short term (1-2 years) the use of impedance based sensors as alternative to LVDTs will be considered as second priority. The method is derived from the concept adopted for the measurement of void formation on the surface of fuel pins in contact with cooling water, but applied to characterize the dimensional changes of the pin itself in dry conditions.

R&D Activity 2: Development of miniaturized, high temperature strain gauges

The objective of this research is to develop sensors that survive the irradiation and thermal conditions of the in-pile environment and are small enough to be inserted (or embedded) in nuclear fuels and material irradiation tests. In order to ensure that the strain gauges are capable of producing high resolution/accurate data from highly curved surfaces in the in-pile environment, this research will conduct investigations in advanced manufacturing, material, and strain gauge theory of the irradiation environment components which are not suitable for current commercial strain gauge technologies. In terms of additive manufacturing, the AJP is highly promising to fabricate arrays of sensors with microscale resolution and tight integration with curved surfaces for high resolution sensing. In contrast to the conventional (e.g. MEMS based) subtractive or semi-additive fabrication methods, the AJP allows 3D conformal printing of a wide range of sensor materials in patterns that complement the surface geometry. The focus of short to mid-term activities (1-3 years) will therefore be the development of printed strain gauges.

In the long term this activity will leverage from the development of piezoelectric materials considered as part of enabling technologies (Section 6.4). Radiation resistant piezoelectric materials could be inserted (or embedded) at fuel and materials interfaces to characterize mechanical stresses (contact pressure). This technology is commercially available for low temperature, non-nuclear applications.

R&D Activity 3: Optical micrometer for in-pile deformation measurement

This activity is a medium to long term project (3-5 years) focused on the development of fiber optic sensors to measure deformation based on Fabry-Perot interferometry, following the successful demonstration of the applicability of the method to in-pile measurement as part of enabling technologies (Section 6.4).

R&D Activity 4: Fiber optic FBG strain sensors.

This activity is a medium to long term project (3-5 years) focused on the development of fiber optic sensors to measure strain based on Fiber Bragg Gratings (FBG) technology, following the successful demonstration of the applicability of the method to in-pile measurement as part of enabling technologies (Section 6.4).

6.2 Materials Properties

Thermal and mechanical properties of nuclear fuel and cladding materials play an important role in determining the operation characteristics of fuel assembles during normal operation and are crucial to understanding the response of fuel during accident conditions. For example, the cladding that surrounds the fuel prevents radioactive fission fragments from escaping into the coolant. In this regard the structural integrity of the cladding, determined by mechanical properties, is of central importance for reducing fuel failure. The cladding must remain ductile and corrosion resistant while maintaining a low absorption cross section for neutrons. In the fuel meat, thermal conductivity is intimately related to energy conversion efficiency as well as reactor safety and is arguably one of the most important material properties. At the atomic scale, degradation of thermal properties is strongly tied to changes in stoichiometry cause by nuclear fission. Larger-scale defects, such as fission gas bubbles and circumferential cracks, also inhibit the transport of thermal energy. Mechanical and thermal properties are strongly influenced by the local environment (e.g. temperature, flux, stress) and can change dramatically over the course of one irradiation cycle.

6.2.1 Thermal Properties

In nuclear fuel, important physical processes such as species diffusion, neutron capture, and microstructure evolution are strong functions of temperature. Thus, prediction of the fuel performance depends on the ability to accurately calculate temperature. Because the temperature distribution within a fuel pellet is largely determined by thermal transport through the fuel assembly, several DOE-NE R&D programs have devoted considerable efforts to understand how radiation damage and fuel burn-up affect thermal transport. Measurements in a post-irradiation environment of *simulation fuel* have provided important new insights into how thermal conductivity changes with burnup. However, the thermal transport of *actual fuel* is intimately tied to the in-pile temperature history, a relation that can only be made based on model predictions. Thus a complete understanding of thermal transport as a function of time-temperature-burnup will require accurate, spatially resolved in-pile thermal transport measurements. Currently the most promising in-pile measurement proposal involves using a standard needle probe to measure thermal transport. It has been found that this approach in its current configuration is not well suited for small diameter fuel pellet geometries.

Alternatively, existing optical and electronic-based thermal conductivity measurement methods have the potential to be adapted for in-pile measurement. The millimeter to micron dimensions of these sensors are ideally suited for small diameter fuel pellets and also provide the potential for local measurement of conductivity (e.g. pellet center vs. high burnup rim structure). However, targeted research needs to be conducted to prove their applicability in extreme radiation and temperature environments. For instance, species diffusion at high temperature may significantly influence the performance of these small sensors. Sensor function may also be distorted by the evolving fuel structure and the unstable fuel/sensor interface. The following discussion identifies key research tasks required to overcome implementation roadblocks and to enable accurate measurement of in-pile thermal transport. There are three R&D activities that are associated with thermal transport measurements:

6.2.1.1 R&D Activities

R&D Activity 1: Thermal conductivity measurement using fiber-based photothermal radiometry

This activity involves the development of a thermal property measurement technique based on photothermal radiometry which will be implemented via optical fiber. The Transient Reactor Test Facility will be the target test reactor for this activity. This choice of test reactor is significant because fiber darkening due to irradiation damage will not come into play for transient and/or short lived steady state tests. An intensity modulated fiber coupled laser will be used to locally heat the sample. Modulated blackbody radiation from the sample will be collected with the same fiber and used to measure the temperature evolution. A continuum based theory will be developed to extract thermal properties from the temperature evolution signal.

By providing local measurement of thermal diffusivity that is commensurate with microstructure heterogeneity, this project will provide significant validation metrics for computation materials science models being developed under the DOE-NE NEAMS program. The small length scale associated with the proposed approach allows the problem geometry to be highly specified, enabling accurate execution of the inverse problem that is used to extract the thermal properties. The small length scale and measurement accuracy of the proposed measurement approach are distinguishing components of this project.

R&D Activity 2: Three-omega sensors for in-pile thermal conductivity measurement and profiling

This activity will involve measuring thermal conductivity of small fuel samples using 3ω sensors (see Appendix A) directly printed onto fuels using additive manufacturing techniques based on aerosol jet technology (see Appendix A). Because fuel cracking and pore migration have deleterious effects for printed sensors, in addition to prototypic fuel pellets we will also consider small (100 μm^3) samples, which have a much smaller thermal stress and are free of pores. The local thermal conductivity along the radius direction of the fuel can be profiled by controlling the sensor heating frequency and corresponding heat penetration depth. In addition, an array of 3ω sensors can be printed along the radius of the fuel pellet to map the fuel thermal conductivity as a function of location.

Device level, multiscale modeling (Appendix A) will be performed to provide detailed insight into 3ω sensors and transduction properties under irradiation. Chemical diffusion at high temperatures and the influence on transduction properties is of particular concern for printed sensors. An additional task under modeling will be directed at understanding the bi-directional relation between electrical resistance and temperature under irradiation. Modeling will also inform the synthesis of new nanomaterial based inks designed to increase the radiation resistance of 3ω sensors, helping to decouple radiation effects from the transduction properties of the sensor.

R&D Activity 3: Needle probes for in-pile thermal conductivity measurement

This focus area will expand upon recent progress in developing a hot wire needle probe (see Appendix A) for in-pile measurements of thermal transport. Initial results indicate the probe design and the thermal contact resistances dominate the temperature response time. Therefore, a significant effort of this activity will be on reducing the thermal inertia and improving the temporal response of current needle probe sensors. This will be accomplished by miniaturizing the probe design with aid of advanced manufacturing techniques, and through the careful selection of materials and their interfaces in order to increase the heat transfer between the probe and the sample. Miniaturization will also have the added benefit of expanding the range of thermal conductivities and small diameter samples which can be measured because the impact of boundary conditions will be reduced as compared to the current design. The miniaturized needle probe design will be optimized using a heat transfer model based on finite element methods, which will also aid in the extraction of thermal conductivity.

Additional efforts in this activity will test the limits of the operational range of the preliminary hot-wire needle probe experiments by expanding the temperature test range and the radiation resistance of the new probe design. It is envisioned that this new probe will target prototypical conditions of real fuel pellets in the ATR. Lastly, we will expand the current measurement scheme to determine the feasibility of performing these measurements in the frequency domain. The diffusion length of the heat pulses can be tuned by varying the frequency of the applied heating pulses. This approach holds the potential to provide details about the radial thermal conductivity profile.

6.2.2 Mechanical Properties

The mechanical properties of cladding materials are strongly tied to evolution of microstructure under irradiation. Corrosion can cause substantial wall thinning and lead to bulging and cladding rupture. Corrosion in the presence of a tensile stress can lead to sudden failure of normally ductile cladding materials. Stress corrosion cracking of materials under irradiation involves very complex failure mechanisms that pose significant challenges to experimental and computational materials scientists. Hydride formation is another important aspect of cladding microstructure that has deleterious effects on mechanical properties. Cracking of brittle hydrides can significantly limit the life of zirconium alloy claddings. The hydrogen required for hydride formation comes from sources such as waterside corrosion, dissolved hydrogen in coolant water and water radiolysis and from internal sources such as moisture an hydrogen that are absorbed in the fuel pellet. Mechanical properties of cladding materials associated with advance reactor designs are also of interest to NE programs. Example materials include, silicon carbide used in TRISO fuel, high chromium content steel cladding materials for fast spectrum reactors and ceramic coatings for advance accident tolerant fuel designs.

6.2.2.1 R&D Activities

R&D Activity 1: Identify path forward

This task will assess potential methods for measurement of mechanical properties in reactor. The analysis will consider which properties of fuel cladding and structural materials are of most interest to the reactor community. Gap analysis will be carried out to identify those areas of engineering and materials science which must be addressed to successfully characterize mechanical properties in reactor. Potential for development of new sensor materials will be determined. Methods to apply and measure loads in the extreme environments of interest will also be addressed. Similar to thermal properties, mechanical properties require measuring the strain of a sample exposed to a well quantified external force. A number of methods for strain determination have been determined and the basics of those methods are described in the appendix to this document.

R&D Activity 2: Bench to demonstration

The methods determined from the analysis carried out during the research plan development will be demonstrated on the bench, for example in a furnace at applicable temperature in the absence of irradiation. Materials science issues relating to development of new sensor materials, e.g., piezoelectrics, will be addressed experimentally. This task will work with researchers from modeling and simulation and advanced manufacturing tracks to coordinate experimental work with their needs.

6.3 Chemistry and Structure

In nuclear fuel, changes in chemistry and structure over the lifetime of the fuel can have a significant impact on fuel performance. In the early stage, radial cracks form due to sever thermal stresses, point defects (Frenkel pairs, fission products) are created, and the stoichiometry starts to change due to fission and subsequent irradiation damage. Also in this stage, the fuel density increases due to pore migration. In the mid stage, point defects start to diffuse; creating voids and causing dislocation growth, fission gas segregates to grain boundaries, and newly created fission gas bubbles start to form. In the late stage, the fuel starts to swell due to bubble nucleation and growth; fission gas is released into the plenum, and the fuel and cladding start to creep. During this stage, swelling within the fuel causes the fuel to come into contact with the cladding. This leads to a chemical interaction between the cladding and fuel that can significantly influence the mechanical properties of the cladding. Continued time in reactor leads to a buildup in the plenum pressure and ultimately to cladding failure. The structural integrity of the cladding is compromised by three primary mechanisms: (1) cladding embrittlement caused by pellet-cladding chemical interaction, (2) cladding embrittlement caused by hydride formation, and (3) wall thinning caused by excessive corrosion.

6.3.1 Chemistry

Chemical evolution in reactor is driven by solid state chemical diffusion which is strongly influenced by the distribution of non-equilibrium defects (i.e. defect distribution that does not exist out-of-reactor). As an example consider oxygen diffusion in oxides fuels. Oxygen diffusion via the interstitial/vacancy mechanism in non-stoichiometric oxide fuel is orders of magnitude faster than its stoichiometric counterpart. Viewed from this perspective it is easy to understand how oxygen diffusion specifically and chemical diffusion in general can be significantly influenced by the non-equilibrium distribution of irradiation-induced defects.

Currently, chemical evolution over the lifetime of the fuel can only be inferred using post-irradiation examination. In light of the discussion above, it is clear that this limitation presents a significant obstacle for achieving a comprehensive and unified understanding of chemical evolution. As an alternative to post-irradiation measurements, we will consider measuring in-pile changes in chemistry in oxides and hydrides using electrochemical impedance spectroscopy (EIS) and high-temperature optical spectroscopy (see Appendix A).

6.3.1.1 R&D Activities

The R&D will move gradually from idealized to prototypic and in-pile testing with some focus, making this transition more quickly. For example, collaboration already is in place between INL and MIT for the study of in-pile cladding hydride formation. Transitioning to study hydride mitigation would set the stage for near-term success. The methodology and the sensor development will be a collaborative effort among researchers in materials development, property modeling, sensor design, and sensor manufacturing. This research activity will encompass three research focus areas aiming at understanding critical challenges facing reactor technologies:

R&D Activity 1: Measuring spatially resolved changes in stoichiometry

This activity addresses the thermodynamic and kinetic variations of oxide fuels as a function of temperature, gas atmosphere and irradiation, and offers in-pile fuel performance information in real-time. The project will deliver a sensor and supporting equipment to monitor and/or measure spatially and temporally resolved fuel stoichiometry. A two pronged approach will be implemented to develop the electrochemical sensor: (1) macroscopically, spatially and time resolved dielectric responses of oxide fuels under thermodynamic equilibria and during transients will be measured at high temperatures under irradiation; and (2) microscopically, advanced tomography, Raman and Synchrotron characterizations as well as finite element modeling will be applied to help interpret the measured dielectric responses so that it is able to pinpoint the physical and chemical properties to be measured. At a later stage, electrode-fuel compatibility, novel sensor design, and advanced manufacturing technique will be added to the electrochemical sensor development to ensure the success and deployment. In FY17, upon the availability of funding, several experimental capabilities will be established, including high temperature EIS under controlled gas atmosphere, APT, Raman Spectroscopy, and Synchrotron XRD. From FY18, we will start the electrochemical sensor development based the above described approach, targeting the dielectric response of oxide fuels under thermodynamic equilibria and during transients. The tasks and milestones will be listed in the corresponding sections.

R&D Activity 2: Monitoring cladding hydride formation and corrosion

This area is a mid-term project aiming at understanding the mechanisms of hydride formation, hydride dealloying and associated cladding corrosion so that a mitigation strategy and technology can be developed for reactor safe operation. The experiments involved include electrochemical measurement of kinetics of hydride formation and hydride dealloying, *in-situ* TEM and tomography characterization of delayed hydride cracking and associated mechanisms as well as visualization of microstructure evolution, synchrotron characterization of the structure of cladding hydride, Raman spectroscopic characterization of hydrogen evolution on surface at elevated temperatures, and so on. At the end of the project, a strategy will be developed and demonstrated to mitigate the hydride formation in cladding. The tasks and milestones are listed in the following sections.

R&D Activity 3: Measuring diffusion of fission gases in oxide fuels

This activity is definitely a long-term project. The diffusion of fission gases depends largely on the evolution of microstructure in fuel pellet and in return influencing the evolution of the microstructure of fuel pellet. Thus far, there doesn't exist any direct experimental observation of fission gas diffusion. We propose to apply electrochemical transient technique to pinpoint the frequency of fission gas diffusion and use segmented measurement method to map the space distributed response during fission gas diffusion in real-time. The success of the project will depend mainly on the determination of fission gas / microstructure sensitive frequency where *in-situ* TEM and synchrotron characterizations will be critical. The goal is to determine the kinetics of fission gas diffusion and the relationship with the evolution of fuel microstructure.

6.3.2 Large-Scale Structure

Large-scale structural changes to the fuel and cladding can a have profound influence on fuel performance and behavior. In some cases such as fuel facture and central void formation, the structural changes can take place in the first few hours after startup. In other cases such as fuel cladding interaction, structural changes may happen over a time span from months to years.

In ceramic fuels, fracturing of the ceramic pellets greatly influences the performance of the fuel. Recent modeling work suggests that substantial radial cracking forms within the first few hours of the initial power ramp. The cracks are formed as a result of high tensile hoop and axial stresses in the peripheral regions of the pellets created by large thermal gradients. As power ramps down, the resulting thermal gradients cause the formation of circumferential cracks. These cracks play a critical role in the fuel performance as they extend the effective diameter of the fuel, closing the fuel-cladding gap, while introducing interfacial thermal resistances lowering the effective thermal conductivity of the fuel. Recent activities in fuel performance modeling have focused on developing mechanistic approaches for predicting fuel fracture behavior in BISON. The effort has resulted in simulation results that produce reasonable cracking profiles. However, the availability of data to validate such detailed model results is limited.

The formation of a central void in MOX fuels is another phenomenon that happens on an hours-to-days time frame following the initial power ramp. Due to the relatively poor thermal conductivity associated with typical oxide fuels, the maximum temperature gradient is in excess of 10^5 K/m. In addition to sever thermal stresses, this gradient as a driving force for atomic transport. The movement of small voids requires transfer of atoms or vacancies form the leading edge to the trailing surface. As these voids move up the thermal gradient, they coalesce in the fuel interior forming a large central void. Fission gas release, swelling, embrittlement, creep and thermal conductivity are strongly related to the motion and growth kinetics of voids. The ability to image the formation of the central void would give computation material scientists unparalleled insight into the kinetics that govern void growth and motion and is of interest to several NE R&D programs.

Additionally, fuel cladding interaction is of great interest for the continued development of both metallic and oxide fuels. In metallic fuels, internal pressure in the fuel pin build rapidly with burnup and resulting fission gas release, while the metallic fuel remains somewhat plastic during fissioning. For modern fuel designs, direct fuel-cladding mechanical interaction (FCMI) is not problematic. However, fuel-cladding chemical interaction (FCCI) is of great importance for predicting fuel performance behavior and is a complex, multicomponent diffusion problem. FCCI results in weakening of cladding and formation of low-melting point compositions in the fuel. The problem is customarily studied by exposing diffusion-couples to high temperatures. In light water reactor oxide fuels, exposure to high pressure water environments causes the cladding to creep down onto the fuel pellet surfaces. Thermal gradients resulting from power increases in the fuel result in fuel fracture as previously described, while fission product accumulation causes fuel swelling. The increase of fuel external diameter and creeped-down cladding becomes critical for predicting fuel behavior. Power increases cause thermal expansion of the pellets, opening of pellet cracks, and release of fission gas that all cause further increase of fuel diameter. The result of these phenomena is the potential for cladding failure stemming from stress corrosion cracking and delayed hydride cracking in the cladding.

6.3.2.1 R&D Activities

R&D Activity 1: Measuring fuel fracture and Central void formation using thermography

This research activity will focus on the application of thermography for the in-pile imaging of nuclear fuels and materials. Infrared thermography (IRT) is a technique in which the thermal radiation emitted from heated objects is detected and used to create an image. Contrast in the image is related to temperature variations across the object. Cracks, voids and discontinuities in a fuel pellet create barriers to thermal conduction and steep temperature gradients which are revealed in the IRT image as shown in Figure 6.1.

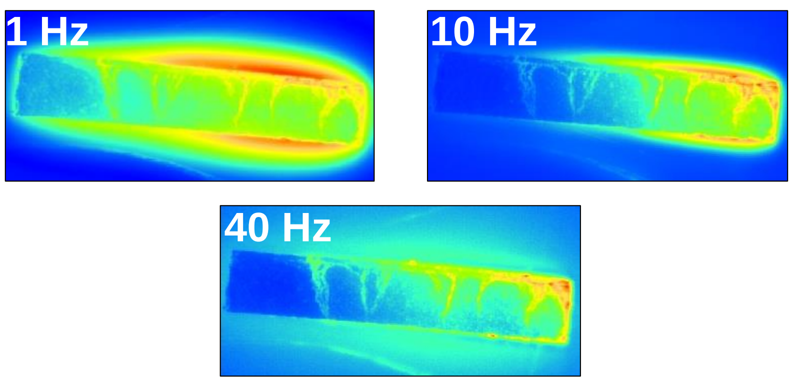

Figure 6.1. Example of lock-in thermography of a cross-section of ZrC taken at different frequencies.

IRT has a number of advantages for in-pile measurements. It is a non-contact technique which requires only optical access to the object. Images can be created in real time and the signal strength increases with increasing temperature. IRT is well developed in commercial industry and has been used for crack detection in high temperature applications. Furthermore, commercial innovations in IRT cameras and detectors can be leveraged for in-reactor measurements.

The primary challenges to the application of IRT for the detection of fuel fracture and central void formation involve the standard in reactor issues including high radiation and also the ability to gain optical access to the sample. Coherent fiber optic bundles, such as those used in some borescopes, can be used to transmit images through pressure boundaries and along curved, non-linear paths. Tasks for this activity will involve the development of a thermographic imaging system which incorporates a coherent fiber optic bundle for image transmission. Proof of principle will be demonstrated by using this imaging system to detect cracks in surrogate samples at elevated temperatures. A capsule which incorporates the IRT imaging system will then be developed for in-reactor testing.

R&D Activity 2: Imaging fuel cladding physical interaction using XRAY

This activity will explore the potential to use x-ray imaging techniques to spatially differentiate fuel from cladding and detect large-scale defects. This activity will address several potential challenges to acquisition, processing and interpretation of image data. Challenges include: characterizing defect types amenable to radiation-based inspection; sensor material survival in a high radiation, high temperature, potentially aqueous environment; selection of radiation source type and spectrum; selection and specification of detector type; configuration of sensor components; compensation for high radiation background; limited view imaging – specification, acquisition, and processing of data. Most of these challenges can be addressed in parallel, however initially emphasis will be place on determining the functionality of detectors in the harsh environment and moving from investigating imaging effects (high radiation background) to investigating environmental effects (high radiation, thermal and fluid).

6.3.3 Microstructure

Microstructure evolution in nuclear fuel is governed by atomic transport facilitated by a highly non-equilibrium distribution of Frenkel defects. With time in reactor, vacancies coalesce forming small voids. Eventually these voids are filled with fission gas. Transport of fission gas bubbles through the fuel to the plenum plays a central role in eventual fuel failure and as a consequence NE funded computational materials science efforts are directed at understanding the mechanisms that govern fission gas transport. Currently there is little effort to conduct bench top studies of fission gas transport in surrogate fuel sample. This is due to difficult of synthesizing surrogate samples with fission gas. Fission gas can be introduced using heavy ion irradiation but the fission gas is implanted only in thin layer, a few microns thick, near the surface of the sample. Monitoring fission gas transport on micron length scales is difficult and extracting the influence of the free surface makes this approach exceedingly difficult. Currently some research groups are investigating novel synthesis methods to entrain fission gas into surrogate samples. Successful fabrication of surrogate sample containing fission gas is a required element to demonstrate bench top instruments capable of measuring fission gas transport.

The interstitial portion of the Frenkel defect is preferentially drawn to dislocation causing dislocations to multiply. The strain energy associated with dislocation production coupled with high temperatures cause profound changes to the grain microstructure. Grain restructuring or recrystallization occurs in both metal and ceramic nuclear fuel. Grain restructuring can result in large changes in texture that will alter both the elastic and plastic properties of fuel. In ceramic fuel, grain refinement in the high burnup rim structure leads to small crystallites having diameters of a few hundred nanometers. In the rim region the large boundary area to volume ratio significantly influences defect accumulation near the boundaries and also impact thermal transport.

Monitoring changes in microstructure of nuclear fuel and materials pose significant technological challenges. While there are many methods to measure and image microstructure in a laboratory setting, few can be extended to an in-pile environment. In this section we consider two broad areas that show promise for in-pile applications. The first area is the application of ultrasonic to measure grain microstructure and the second is the extension of techniques that employ electromagnetic radiation to characterize microstructure.

6.3.3.1 Microstructure Characterization using Ultrasound

Ultrasonics holds great potential to indirectly measure microstructure in an in-pile environment. This approach involves relating the constitutive relation between stress and strain to microstructure. A typical experiment measures either ultrasonic velocity or attenuation. For instance, measuring the resonant frequency of a long narrow bar will give an expression of the modulus of elasticity, E. Measurement of the bulk longitudinal velocity will give an expression that contains E and Poisson's ratio, ν. For nonattenuative, isotropic materials, these two elastic constants serve to define the complete stress-strain relationship. However, all materials exhibit attenuation and are rarely isotropic. Departure from isotropy requires more elastic constants to define the stress-strain relation. For instance, a polycrystalline metal plate that has been textured by rolling requires nine elastic constants to define the stress-strain relation. For attenuative materials involving the conversion of ultrasonic energy into another form of energy, the ultrasonic velocities, in general, are functions of frequency. In this case, the relation between stress and strain is referred to as viscoelasticity.

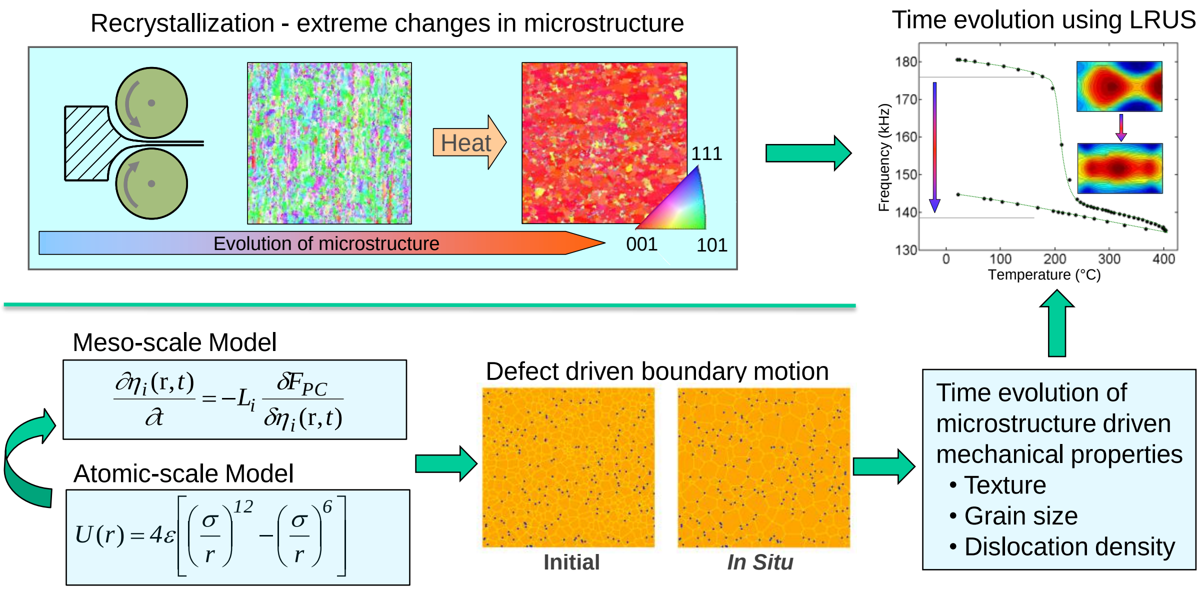

Figure 6.2. Recrystallization of heavily deformed high purity copper. RUS is used to monitor the polycrystalline elastic stiffness tensor. A parallel mesoscale modeling effort will be used simulation the time evolution of the microstructure.

Large microstructure features, like voids/pores, have a strong influence on ultrasonic velocity. This has lead researchers to explore the relation of changes in velocity to changes in swelling in thick-walled reactor vessel components. This characterization approach has the added advantage of being able to spatially map variation in swelling. Changes in texture caused by deformation-induced recrystallization can also dramatically influence velocity. Similar to deformation-driven processes, irradiation-induced dislocation growth causes nuclear fuels to recrystallize and/or restructure. Ultrasonic attenuation can also be related to changes in microstructure. Using Granato Lucke theory, ultrasonic attenuation can be related to dislocation line length and density.

In principle, the concept of relating constitutive relations to microstructure is easy to comprehend; however, in practice there are several issues that need to be addressed to accurately and uniquely make this connection. First the size of the effect must be considered. For instance attenuation due to irradiation induced-dislocation growth can be orders of magnitude smaller than attenuation caused by slight changes in texture. Thus, accurately associating changes in dislocation density and pinning length to ultrasonic attenuation will require the use of single crystal samples. Attenuation due to ultrasonic coupling to the environment can also overwhelm the contribution from dislocations. Accounting for this source of apparent attenuation requires designing experiments where the sample is ultrasonically isolated from the environment. Changing coupling conditions associated with contract transduction methods can also lead to issues associated with measurement reproducibility.

Other issues are inherent to ultrasonics and are related to the physics of wave mechanics. All ultrasonic waves experience diffraction or spreading of the wavefront. This gives rise to another source of attenuation called geometric attenuation. Correctly accounting for this source of attenuation requires confining the ultrasonic energy to a particular geometry. This approach, termed ultrasonic resonant spectroscopy, involves measuring the mechanical resonances of an ultrasonically isolated sample. Another issue related to wave mechanics is referred to as frequency-dependent dispersion. For wave-guiding geometries, such as long narrow bars or plates, the ultrasonic velocity is a strong function of wavelength when the wavelength is comparable to the sample dimensions. Wave-guiding geometries can be readily modeled; however, this does add another layer of complication that should be avoided if possible.

R&D Activity 1: Resonant Ultrasound Spectroscopy to Monitor Recrystallization

This R&D activity will involve using ultrasonic techniques to measure grain restructuring in metallic fuels. The approach will involve relating changes in polycrystalline elastic properties to grain microstructure. Resonant ultrasonics is the technique of choice for two reasons. First, large amplitudes can be obtained with small forcing functions. This is particularly important for experimental methods that employ radiation hardened piezoelectrics that typically have small piezoelectric coupling coefficients. Second, resonant techniques offer the highest absolute accuracy of any routine elastic modulus measurement technique.

There are two important tasks associated with this R&D activity. The first involves designing the instrument that will be used to excite and measure resonant frequencies. Piezoelectric, EMAT and laser transduction will be considered. A key component of this task will be sample synthesis and sample holding. For sample synthesis it is important to obtain a starting grain microstructure that will evolve in a controlled and predictable fashion under irradiation at high temperature. It is also important to hold the sample in such a way to limit ultrasonic coupling to the environment. This is especially important for measurements of ultrasonic attenuation.

The second major task will be modeling the expected response of the sample under irradiation. Defining an envelope of expected outcome is critical to optimizing instrument design. The modeling approach will involve developing a phase field model of recrystallization that accounts for dislocation density. Coupled modeling of deformation and recrystallization is a hard problem. It requires the generalization of the model summarized in to finite deformation and then couple the deformation outcome to the recrystallization model. Initially this task will be limited to demonstrating a first connection between the deformation and recrystallization models by transferring the deformation data to a simplified recrystallization model based on the phase field approach. The goal is to demonstrate that a detailed representation of the internal energy distribution and internal stress evolution yield results that are different from those obtained from current models starting from the same microstructure but without including the detailed distribution of the dislocation density and internal elastic energy.

6.3.3.2 Microstructure Characterization using Optics

Two promising optical-based measurement methods that have the potential for in-pile operation are Raman spectroscopy and optical microscopy. These characterization techniques can provide information on microstructure and chemical changes that occur during irradiation. Structural and phase changes can result in decreased fuel performance, potential hot spots, stress concentration, lower mechanical properties, irradiation growth, lower melting points and embrittlement due to chemical segregation. Optical microscopy can provide information on porosity or crack formation and Raman spectroscopy can provide information on local chemistry and structural.

The initial effort will involve the development of a free-space optical beam delivery system, using steering mirrors, which will be less susceptible to radiation-induced degradation of optical components. It is envisioned that the free-space optical system could be used for both optical microscopy and Raman spectroscopy. There are many challenges that must be addressed to realize in-pile measurement of microstructure using optics-based methods:

- Mechanical vibrations can cause optical misalignment
- Configuration and size limitations of the access portals that can be used for feeding the laser source
- Loss of intensity or scattering of signals can cause loss or incorrect collection of data
- Only measure near surface microstructure
- Environmentally induced degradation of mirror/sample reflectivity
- Sample size and accessibility
- Both Raman spectroscopy and optical microscopy rely on properly prepared samples. Degradation of surfaces in a reactor environment is a critical issue

6.3.3.3 R&D Activities

R&D Activity 1: Raman Spectroscopy to identify structural and phase changes associated with temperature and fission product generation.

Initially, the emphasis will be on development of Raman spectroscopy capability. The assumption is that if successful, the optical system for Raman can be extended to optical microscopy. Raman spectroscopy is an effective contactless technique suitable for structural and chemical characterization of ceramic nuclear materials, such as uranium dioxide (UO_2), thorium dioxide (ThO_2) or cerium dioxide (CeO_2). Vibrational spectra can provide information on phases and phase transformations (e.g. identifying different phases of uranium oxides), as well as defects and oxygen stoichiometry. Raman spectroscopy can also be used for quantitative compositional analysis of mixed oxides (e.g. UO_2-ThO_2), and for probing changes in the vibrational spectra caused by external factors, such as temperature, pressure, irradiation, corrosion, etc. Additionally, Raman can be used to determine the oxidation states of oxidized fuel or cladding material, and ceramic cladding materials like SiC or SiC-graphite composite materials.

In situ characterization of nuclear fuel materials inside of a reactor, implies spectral measurements at high temperatures. Ultraviolet (UV) lasers, such as 325 nm line of He-Cd laser or 363.8 nm line of Ar ion laser will be used for excitation of Raman spectra. UV excitation presents an important advantage over visible light for the proposed tasks. A difficulty in measuring Raman spectra of materials at high temperatures using visible excitation is caused by continuous background due to thermal emission, which can be rather intense compared to weak and broadened Raman features. UV excitation shifts Raman spectra to much shorter wavelength range, far away from the peak intensity of thermal radiation. It was demonstrated that in the case of conventional visible excitation using green lasers, thermal radiation background becomes significant at temperatures of 800–900°C, and rapidly increases at higher temperatures, while the UV excitation (325 or 363.8 nm) allows measuring Raman spectra with practically no backgrounds up to as high as 1800°C. Another potential advantage of using UV excitation results from the ω^4 dependence of the Raman scattering cross section, which should lead to an increased intensity of the Raman signal, which may be essential for the proposed experiments.

The use of a reflective optical system for laser delivery will be investigated. A reflective optical system may obviate replacing a fiber feed delivery system for monitoring, thereby enabling more continuous monitoring. The optical system is an enabling system for Raman spectroscopy that must operate in extreme radiation and temperature environments. Additionally, optical misalignment due to mechanical vibrations will have to be addressed. Early phases will include system specification, materials selection and opto-mechanical and structural engineering. The later phases will include implementation of the system including risk mitigating experimental studies, all with the goal of developing a successfully functioning system. Assessment of all risk factors for the lifetime of the system, temperature cycling and associated factors and radiation exposure are integral to the study as key risk factors differentiating this optical system from ordinary applications in less harsh environments.

For this specific study, the research activities will focus on the structural and phase changes associated with temperature and fission product generation and interactions. In situ Raman spectra will be used for probing the structural evolution of nuclear ceramic materials under extreme conditions (temperature and radiation), identification of composition and phase changes and structural defects leading to the appearance of defect-induced Raman bands. The spatial resolution and sensitivity of Raman spectroscopy can potentially allow for characterization of the sample inhomogeneity with resolution of few hundred µm, and measurements of kinetics with a time resolution of several seconds to minutes.

6.3.3.4 Microstructure Characterization using X-ray Diffraction

There are a host of lab-based instruments that can be used to characterize microstructure. Extending these to an in-pile environment poses significant challenges. However, both physical and administrative accessibility requirements at research reactors around the world are being lowered. It is thus an opportune time to consider extending traditional methods for characterizing microstructure to in-pile environments. In this section of the program plan we discuss one such technique that may have application for in-pile measurements. X-ray diffraction (XRD) is a technique that provide information on the atomic structure of the material, the chemical bonds, and indications of chemical interactions as well as elastic properties of materials. XRD spectroscopy analyzes both the wave and particle properties of X-rays to acquire information about the structure of the evaluated crystalline material. The main use of this technique is to identify and characterize compounds based on their specific diffraction pattern and is unique for each specie, phase or oxidation type.

Technique description

The X-ray diffraction patterns (reflections) are produced when the crystal is illuminated by the X-ray and providing a 3D projection if these patterns are collected at different angles. The scattered X-rays undergo destructive and constructive interference upon impact. The diffraction of X-rays can be described by Bragg's Law where:

$$n\lambda = 2d\ sin\theta$$

where *n* is a positive integer, λ is the wavelength, *d* is the lattice spacing and θ is the angle of incidence.

The direction of the scattered X-rays strictly depends on the size and shape of the unit cell of the target material. The intensity of the diffracted waves is affected by the atomic arrangement in the crystal structure. The diffraction pattern will be unique for every atom, allowing to identify the composition of the target material. Figure 1 displays the schematics of the X-ray diffraction technique.

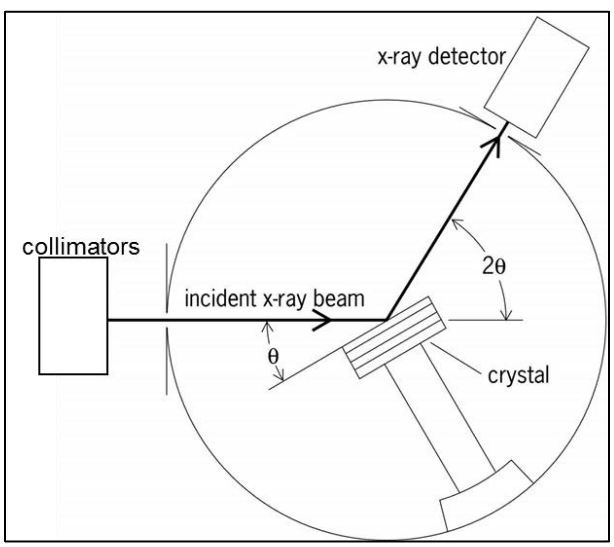

Figure 6.3. Schematic of X-ray diffraction.

Free electron lasers have been developed for use in X-ray crystallography with the X-rays coming in femtosecond bursts. The intensity of the source is such that atomic resolution diffraction patterns can be resolved for crystals otherwise too small for collection. However, the intense light source also destroys the sample requiring multiple crystals to be shot. As each crystal is randomly oriented in the beam, hundreds of thousands of individual diffraction images must be collected in order to get a complete data-set.

A pinhole collimator is used to control the beam size and divergence. In an XRD system, the pinhole collimator is normally used together with a monochromator or a set of cross-coupled Göbel mirrors. The beam divergency decreases continuously with decreasing pinhole size for the combination of double pinhole collimator and monochromator. For quantitative analysis, texture, or percent crystallinity measurements, 0.5 mm or 0.8 mm collimators are typically used. In the case of quantitative analysis and texture measurements, using too small a collimator can actually be a detriment, causing poor statistical grain sampling. In such cases, statistics can be improved by oscillating the sample.

A collimator is an aid that helps to reduce the net cross section of an X-ray beam. A collimator is made of Pb- or W- alloy or other strong attenuating material. The aperture is either a 1D slit or a 2D aperture having circular, quadratic, rectangular or other shape. The collimator resolution Rx (and Ry) is the nominal width of the aperture in the appropriate direction. The x-ray throughput passing the aperture thus is proportional to Rx and Ry. The throughput or collimator efficiency g as $g \sim Rx*Ry$ or $g \sim R^2$ in the case of a rectangular or circular aperture of the collimator. Collimators have to be used in XRD (as well as in HRXRD) to define the Bragg angle and the angular resolution. They are used in many other x-ray applications for example reducing the x-ray dose to patients or reducing x-ray scatter impinging on a detector.

The radiation produced consists in rays traveling in a variety of directions and consisting of a spread of wavelengths. The purpose of the collimation portion of an XRD instrument is to produce a relatively thin beam of X-rays with a narrow spread of wavelengths, all traveling in essentially the same direction. Some commonly used components are described below.

- **Slits or pinholes**: These form a part of almost every instrument, and act by geometrically restricting the beam. To be effective, they must be constructed from a heavy element such as tungsten. Care must be taken to minimize diffuse ("parasitic") scattering from the edges of the slits which can contribute to the background signal.
- **Crystal monochromator**: The most common method for producing a "monochromatic" beam (containing only a narrow spread of wavelengths λ) is to insert a high quality single crystal of a material such as silicon or germanium into the beam and separate out only those components of the beam that satisfy Bragg's Law. Conversely, for a beam that is already largely monochromatic, this Bragg reflection from a crystal can be used as a means of collimation. The degree of collimation and spectral selection depend on the perfection of the crystal and also the characteristics of the incoming beam.
- **X-ray Mirror**: X-ray mirrors consist of a beam which strikes a flat surface at a very low angle and can be strongly reflected. X-ray mirrors are typically made of a metal such as gold and are gently curved so as to produce a beam that is focused along a vertical and/or horizontal axis. They also affect the spectral characteristics since shorter wavelengths are reflected much less effectively than long wavelengths.
- **Multilayer Optics**: This approach, which is incorporated in many units currently on the market (especially those optimized for small-angle scattering) combines the benefits of a crystal mono-chromator and an X-ray mirror. A multilayer coating on a curved substrate results in a

monochromatic, collimated beam, most often either parallel or slightly convergent focus. The optical unit must be closely coupled with the source, but when done properly this can result in a beam that is simultaneously more intense and better collimated than achievable with previous technologies.

Challenges

Although the benefits by having XRD data available as reactions occurred during irradiation, especially to identify any transient and/or unstable phases that formed during the process of irradiation and temperature in the presence of fission products, multiple challenges are known that may impact the feasibility to implement XRD as an in-situ and in-pile measurement technique. It is therefore the scope of the initial research phase, to evaluate the feasibility of the XRD measurement technique in an in-pile environment. Listed below are a preliminary list of potential challenges that can hamper the feasibility for measurement execution. As XRD is a measurement technique that can be applied for multiple material types, and not only limited to mostly ceramics, it is a versatile technique that can provide many benefits as a research tool, and a validation tool for some of the other indirect techniques. This is one of the reasons that the initial challenges, should not be self-limited until such time that a detailed feasibility study with recommended solutions are proposed or developed. Some of the challenges, may be similar to those identified during the initial research plan development for Raman measurements. We recommend thus that XRD research activities initially include the feasibility study to identify any other potential challenges unique to XRD measurements, and to benefit from the lessons learned during the initial work on Raman experimental evaluations that can be related to similar behavior for XRD evaluations.

- Effect of irradiation on the characteristics of the incoming beam
- Collimator:
  - Size: Using too small collimator can cause poor statistical grain sampling. In such cases, statistics can be improved by oscillating the sample. (in the case of powder samples)
  - Distance of collimator from sample if chosen to have collimators outside the reactor chambers (like with potential Raman measurement designs). Generally, the optical unit needs to be closely coupled with the source
  - If beam is too focused, can damage sample or otherwise too much scattering occur if it is defocused
  - Material type of collimator: effect of irradiation and temperature on collimator if the in-pile system design require close approximity. (Generally a collimator is made of Pb- or W- alloy or other strong attenuating material).
  - Effect of irradiation , temperature on the slit opening: prevention of scattering
- Beam too intense and will “burn” the sample or if the optical unit is not close enough to the source, the beam is not focused and loss of resolution or no measurement.
- Mirrors: Similar challenges as identified for the Raman measurement technique. The lifetime and accuracy of measurements may be influence by the:
  - mirror material degradation due to irradiation
  - optical lens material and behavior which needs to be evaluated over time and conditions. Darkening or microstructural changes may influence accuracy and resolution

R&D Activity 1: X-ray diffraction (XRD) to determine structure and mechanical properties

The efficacy of making in-pile XRD measurements will carefully addressed. A set of nuclear materials will be identified that are best suited for X-ray studies. This set will span from single phase, single crystals to multiple phase polycrystalline samples. A key element of this scope will be understanding the challenges associated with this type of measurement.

6.4 Enabling technologies for advanced sensors

This program has identified advanced manufacturing processes as a potentially disruptive technology to develop innovative instruments for in-pile test of nuclear fuel and materials. Other enabling technologies have a similar potential to provide a substantial contribution to advancing the performance of a wide number of instrumentation activities. In this section innovative technologies having longer-term goals are considered, based on three classes of advanced materials: thermo-electric materials for power harvesting, piezoelectric materials for wireless transmission and beyond state-of-the-art optical fiber materials for long term application in radiation environments. These low maturity technologies address the common challenge posed by feeding power and signal transmission cabling into the core of nuclear reactors. In addition to issues related to the complexity of design and implications to safety, metal based cabling are subject to high level of noise and interference. Optical fibers eliminate the issue of electronic noise and allow the deployment of distributed sensors providing multiple, local readings. In addition, development of new optical fiber materials maybe instrumental to the long-term deployment of several advanced measurement techniques described in the previous sections. In some cases, self-powered detectors coupled with wireless data transmission would eliminate the need of feeds altogether, providing a breakthrough for the deployment of sensors in test reactors.

In the short term the research activities considered will be focused on the identification, fabrication and validation of innovative materials. The material science thrust is therefore expected to play a fundamental role, in particular the application of High-Throughput Computational Screening (IHTCS) methods.

6.4.1 Radiation tolerant optical fibers

The development of radiation tolerant optical fiber is a crucial step in enabling innovative solutions for measurement of the field and material properties discussed in previous sections of this document. The activities will support in particular the following:

- Temperature measurement
- Fission gas pressure and composition measurement
- Thermal conductivity measurement and thermography measurements
- Deformation measurement

R&D Activity 1: Radiation Tolerant Pressure Seal for Optical Fibers

The first research area is focused on development of an enabling technology required for any in-situ fiber optic based instrumentation. Sensors installed within a test fuel rod must pass through a pressure boundary. Internal fuel rod pressures may exceed 5 MPa. The sensor most commonly installed in this fashion is a thermocouple, which is effectively a metal sheathed solid rod. As such, thermocouples are typically installed via brazing or through a Swagelock style compression fitting. Neither of these methods are appropriate for a fiber based sensor. To deploy a fiber based sensor in a fuel rod a new high temperature, radiation tolerant seal must be developed.

R&D Activity 2: Develop radiation resistant optical fiber materials for in-pile application.

There are several groups, both nationally and internationally that are conducting research on radiation resistance fibers for nuclear applications. According, the first effort is to perform a survey of the state of the art with respect to radiation tolerant optical fibers and associated measurement techniques. This will include a gap analysis identifying needs for newer materials and sensors to be developed as part of this program. The gap analysis will also identify infrastructure needed to perform these tasks and outside organizations that could be brought in as collaborators.

In this activity, models will be established to look into various materials that can serve as potential materials for fiber or as dopants to conventional silica for fiber. Prior research has established various approaches to improve fiber based sensors by changing the material of the fiber, including ongoing research as part of NE programs on sapphire and concepts based on SiC. The modeling will also look into the optical and sensor characteristics of the identified materials. Additionally, novel materials will be modeled and identified that can enable sensor fabrication using additive manufacturing. For example, metal nanoparticles with Si have been demonstrated to make a new form of grating. Such nanomaterials can be deposited using additive manufacturing techniques, such as ink-jet or aerosol jet printing.

R&D Activity 3: Design and Fabrication of PCFs for in-pile sensing.

In this focus area, Photonic Crystal Fibers (PCFs) will be investigated for in-pile sensing. PCFs can be designed to have a high wavelength dependent dispersion and a refractive index profile that is sensitive to temperature. It can serve as an ideal temperature sensor depending on the material used to make the PCF. Additionally, PCPs may be the fiber material of choice for thermography and/or thermal conductivity measurements.

6.4.2 Power Harvesting

The power harvesting technology has crosscutting significance to enable self-powered wireless operation and thus address a critical technology gap for in-pile sensors and instrumentation. Efficient and reliable power harvesting technologies not only significantly expands in-pile sensing and instrumentation capabilities but also offer major cost savings over current approaches requiring cable installation and external power sources. In addition, the self-powered wireless sensor networks could support the long-term and safe monitoring of the new reactor designs and fuel cycle concepts. Several important issues need to be addressed to realize successful deployment of in-pile power harvesting technologies. First, the power harvester needs to be placed within reasonable proximity to the sensor while producing sufficient power density in a given space to power both the sensors and wireless transceivers (assuming two way communications). Second, the power harvester needs to operate reliably under the radiation and high temperature in-pile environment. Third, the power harvester needs to be compact and scalable to fit the in-pile space constraints and effectively utilize the available energy sources.

R&D Activity 1: Develop thermoelectric generator system for in-pile power harvesting

The research goal of this activity is to develop and deploy robust and efficient thermoelectric generators (TEGs) that can harvest thermal energy in nuclear reactor core and enable self-powered wireless sensors. This will include Thermally Chargeable Solid-state Supercapacitors made from a solid-state polymer electrolyte. Among all the potential power harvesting technologies, thermoelectric power harvesting is a promising power harvesting technology for in-pile applications due to its compact and solid-state nature and the large temperature gradient available from the fuel core to surrounding coolant. This activity will focus on three research objectives: (1) Identify irradiation-resistant and efficient thermoelectric materials; (2) Design and fabricate compact and robust TEGs using advanced manufacturing method; (3) Deploy and demonstrate a self-powered sensor system by integrating the power harvester with in-pile sensors. Several innovative approaches will be utilized to achieve our research goal. In the short term (1-2 years), combinatorial high-throughput materials science and workflow will be developed for radiation-resistant thermoelectric materials discovery. In the mid to long term (3-5 years), advanced manufacturing method will be developed to fabricate robust and compact power-harvesting devices that can be integrated with fuel systems. In addition, nanostructured thermoelectric materials will be explored in order to increase the efficiency in converting heat into electricity, and improve radiation resistance over conventional thermoelectric materials.

Other technologies to harvest power in the core of nuclear reactors will be considered based on the results of activity 1 and preliminary screening of performance and deployment feasibility. A detailed work plan will be developed as part of the research activities. The main technologies considered, in addition to the TEGs described above, are:

- Self-Powered Neutron Detectors (SPND) as power source
- Inductive and ultrasonic coupling
- Power Harvesting Using Mechanical Vibrations
- Ultrasonic transmission

6.4.3 Data transmission and analysis

Data transmission presents a major challenge for in-pile sensors and instrumentation, as discussed in the introduction to this section. Traditional wired signal transmission and communication is difficult and expensive to implement in-pile due to design and safety constraints and the very harsh environment, both in terms of material degradation due to neutron damage and noise interference from operating components. Wireless transmission has the potential to provide breakthrough advancements in the deployment of in-pile sensors. This potential would be enhanced by the coupled development of self-powered sensor, in order to eliminate the need of metallic cabling altogether. However, wireless data transmission alone would be a significant enhancement to sensor deployment even if coupled with wired power lines.

Conventional wireless communications systems based on narrowband and spread spectrum signals face significant challenges in nuclear environments. In-pile communication must transmit signals through reactor pressure vessels and concrete walls as well as manage signal deterioration as the communication system performance decreases as a consequence of material degradation under irradiation. The short term task of the activities is therefore to develop innovative materials for radiation tolerant signal transducers. A second near term task is to improve the performance of wired sensor under development by understanding and controlling operational noise in irradiation facilities considered for deployment. These activities will provide feedback to optimize instrumentation design from the perspective of noise reduction, as well as guide the development of appropriate data acquisition system (DAS) for all sensors considered for deployment. The work plan for the activities in this area will be developed as part of the research activities.

Another active area of research is the development of data analytic tools with general applicability to instrumentation under development as part of this program. Two main challenges need to be considered in this area as part of the deployment of instrumentation in irradiation test: the storage of large amount of data inherent in many of the technologies under development giving the constraints on physical access and data transmission during operation; the integration of specific sensor signals with the data collected as part of by the irradiation facility operation, which is essential to efficiently process and evaluate the sensor performance.

R&D Activity 1 Piezoelectric based acoustic transducers for in-pile wireless communication

The research goal is to develop high-temperature and radiation-resistant piezoelectric materials and transducers for in-pile signal transmission and wireless communication. In the short term (1-2 years) the result of combinatorial high-throughput screening analysis discussed in the previous section will be leveraged to identify innovative piezoelectric materials. Magnetostrictive materials used as part of Ultrasound Thermometers (UT) development (Section 6.2) will be considered as alternative candidates. In the mid to long term (3-5 years)advanced manufacturing methods will be developed to fabricate robust and compact piezoelectric acoustic transducers that can be integrated with in-pile systems. It should be noted that the development of radiation resistant piezoelectric transducers would impact several other activities considered as part of this program, in particular for the measurement of field properties.

R&D Activity 2 Electromagnetic interference (noise) suppression for in-pile instrumentation

In the short-term (1-2 years) the objective of research activities is to analyze the data from sensor deployed in irradiation facilities considered as part of this program in order to define a set of reference signals caused by electromagnetic (EM) interference. These digitized noise signatures will be used in anechoic chambers in order to characterize the performance of sensors under development in terms of noise control. The development of a standard testing process will aid comparative analysis and guide the sensor and data acquisition system (DAS) design optimization for field properties instrumentation. In the mid to long term the activities will consider the effect of EM noise on innovative data transmission system developed as part of this program, such as radiation resistant wireless transducers, and contribute to the deployment in irradiation facilities of novel characterization techniques for material properties and structure discussed in previous sections.

R&D Activity 3 Development of data analytic tools for in-pile instrumentation

The objective of this activity is to develop a common data analytics software tool for the signals transmitted by the instrumentation developed under this program during in-pile testing. The tool will address the following 3 phases: data storage, analysis and reporting. In the near term (1-2 years) we will identify the most efficient combination of commercial tools and customized platform based on the requirements of field properties instrumentation considered for first deployment, in particular in the TREAT facility. In the mid to long term (3-5 years) the tool will be progressively extended to include capabilities required by all other sensors planned for deployment in irradiation tests.

In addition to piezoelectric transducers considered in Activity 1, other technologies for wireless signal transmission from the core of nuclear reactors will be considered based on the results of material development and preliminary screening of performance and deployment feasibility. A detailed work plan will be developed as part of the research activities. The main technologies considered are:

- Adapting Traditional RF Communication for In-pile Environment
- Ultra-wideband Communication for In-pile Environment
- Inductive coupling

7. CONCLUSION

This vision of these advancements in in-pile instrumentation and characterization will require breakthroughs in several key areas, including modeling and simulation, material science, and manufacturing for sensors development. They directly correlate to material performance with real-time evolution of microstructure during irradiation. This relationship can only be inferred today through the examination of materials post-irradiation.

Advancements in measurement and characterization capabilities will support a variety of needs shared by a number of NE programs. Sensor development will initiate observations of real-time behavior of fuel and materials, provide the opportunity to improve measurements of material behavior during irradiation, improve sensor coverage in irradiation experiments, supply smaller length-scale data to understand materials changes due to irradiation, provide data supporting modeling and simulation on a microscopic scale, and provide expertise gathered from research on irradiation-exposure durations.

By addressing the needs and grand challenges identified by existing R&D programs, capabilities will be created in materials science, modeling and simulation, manufacturing, and sensors design and development that will enable breakthroughs for the design of future instrumentation for other missions that will be required by NE R&D programs. This includes (1) the ability to measure and monitor critical process parameters in advanced reactor and fuel cycle facilities, (2) technologies needed for the control of nuclear energy processes and systems, (3) communications technologies for collecting, storing, and securely transmitting data in energy system networks, and (4) technologies that can be used for measuring and monitoring systems in beyond-design-basis events. They will also provide the basis for delivering unique sensors to support requests from commercial fuel and energy system vendors through the GAIN initiative, to utilize irradiation facilities for fuels and materials studies in next generation nuclear energy system design and qualification.

The improvements described here can be achieved by employing innovative tools and methods that reduce technology-development costs, cycle times, and program risks. Improved material screening and selection is expected to optimize sensor design prior to laboratory construction and testing. In addition, the reduced costs and improved likelihoods of successful sensor development strategically align with the mission goals of Gateway for Accelerated Innovation in Nuclear (GAIN) by providing innovative in-core measurement capabilities that are needed to supplement the irradiation testing, validation, and commercialization of new nuclear fuels and materials under development by the nuclear community. These capabilities will be a tool for users of the R&D test bed by allowing for rapid and cost-effective measurement of material properties needed to achieve and advance the mission of the DOE-NE.

8. REFERENCES

A comprehensive list of references is in progress and will be made available for the final draft of the Technical Program Plan.

Appendix A

Measurements and Methods

Appendix A
Measurements and Methods

Three omega measurement of thermal conductivity

The three omega technique employs a small heater wire that is applied to the sample of interest. The heater wire is driven by an 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 AC 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 can be measured using a lock-in amplifier. The 3ω thermal conductivity measurement method is advantageous for thermal conductivity measurement at high temperatures due to the minimized radiation influences. The modulation heating provides effective control of the heat penetration depth in the sample. The thermal penetration can be confined close to the heater region so that the edge effects due to finite sample sizes can be eliminated.

Multiscale modeling methods

The methods are based on phase field and finite element modeling, implemented by MOOSE, to simulate the sensors. Their modeling input parameters, such as materials properties / diffusivities / conductivities, can be estimated using the smaller length scale modeling methods (DFT or MD)

Hot-wire needle probe thermometry

The hot-wire needle probe utilizes the thermal resistance environment of a heat source embedded in a medium to probe the medium's thermal conductivity. The hot-wire needle probe uses Joule heating to create a condition of thermal equilibrium and heat the sample with a constant power. The temperature rise is measured using a co-located thermocouple and monitored as a function of time. If the power is generated through a pulse of current, then a plot of the transient behavior of the temperature rise can be used to extract the thermal conductivity of the medium. The thermal conductivity is given as:

$$k = \frac{CQ}{4\pi LS} \quad (1)$$

where, κ is the thermal conductivity, C is a calibration factor for the probe, Q is the power dissipated by the heater, L is the length of the heater, and S the slope of the linear portion of the temporal response given by:

$$S = \frac{T_2 - T_1}{ln\frac{t_2}{t_1}} \quad (2)$$

Here, *T* is temperature and *t* is time. The calibration factor *C*, can be ignored if the probe diameter is miniaturized below 2.5 mm, while L and Q can be engineered to increase probe sensitivity and tailor the probe response time.

Preliminary experiments have revealed several insights regarding in-pile applications of the hot-wire needle probe. The thermal inertia appears to be dominated by the thermal contact between the probe and the sample. Smaller diameter samples and high thermal conductivity samples limit the time available for testing with large probes. Thermal boundary conditions and probe design can be engineered in order to enable rapid and accurate evaluation of thermal conductivity.

Fiber-Based Photothermal radiometry

Photothermal radiometry is a versatile measurement technique and can be implemented via optical fiber. This technique uses an intensity modulated laser to heat the sample and the resulting increase in infra-red (IR) thermal radiation is detected and analyzed to determine the thermal properties. A unique aspect of this approach is that the laser beam is delivered and the IR signal returned via the same optical fiber. The modulated laser beam is focused onto the sample resulting in an increase in the temperature of the sample at the focused spot. The increase in temperature results in an increase in the IR radiation emitted by the sample. When the modulated laser beam is off, the sample cools by conducting the heat away and the thermal radiation emitted by the sample decreases. The shape of this modulated IR signal depends on the thermal properties of the sample. These properties can then be obtained by comparing the system response to an existing continuum based theory.

Compared to traditional methods for thermal diffusivity measurement, this technique has the advantage of high portability and high spatial resolution. Measurements can be made in a matter of seconds and the measurement length scale is in the 20-200 µm range, which means the thermal properties are measured on a scale much smaller than the microstructure heterogeneity within a fuel sample. The signal using the photothermal technique increases as the temperature increases and sample coating is not an issue, so it is well suited for high temperature measurements. Furthermore, since it is non-contacting, the fuel or sample geometry does not need to be altered to accommodate a sensor.

Linear Variable Differential Transformers

The creep test systems in the lab here at Idaho National Laboratory utilize a linear variable differential transformer (LVDT) device to measure a specimen’s changing dimensions during testing. These devices are electrical transformers which measure linear displacement by converting a position from a mechanical reference position into a proportional electrical signal containing both phase, to represent direction, and amplitude, to represent distance, information. LVDTs do this through electromagnetic coupling, utilizing three solenoidal coils placed end-to-end around a tube (figure 1).

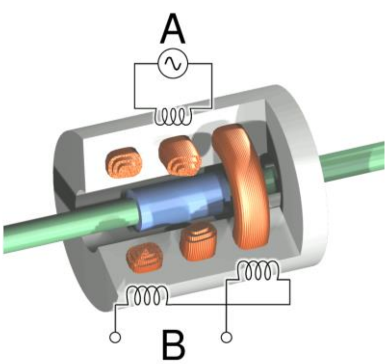

Figure A-1. Linear Variable Differential Transformer [1]

The center coil (A) is the primary, and the two outer coils (B) are the top and bottom secondaries. A cylindrical ferromagnetic core, attached to the specimen to be measured, slides along the axis of the tube. An alternating current drives the primary and causes a voltage to be induced in each secondary proportional to the length of the core linking to the secondary. As the core moves, the primary's linkage to the two secondary coils changes, and causes the induced voltages to change. The coils are connected so that the output voltage is the difference between the top secondary voltage and the bottom secondary voltage. When the core is in its central position, equal voltages are induced in the two secondary coils which cancel, giving an output voltage of zero. When the core is displaced, voltages are induced in the secondary coils, resulting in an output voltage greater than zero which is either in phase with the primary or opposite to that of the primary voltage. The synchronous detector can then determine a signed output voltage that relates to the displacement of the specimen [1].

HEIDENHAIN Length Gauges

Some of the newer creep test systems utilize HEIDENHAIN length gauges, which utilize an incremental measuring method in conjunction with a photoelectrical scanning principle to measure a distance. With the incremental measuring method, the position information is obtained by counting the individual increments from an absolute reference point. The HEIDENHAIN encoders operate using the principle of photoelectric scanning which, depending on the size of the grating period, utilizes either the imaging principle, which functions by means of projected light signal generation, where two scale gratings with equal or similar grating periods are moved relative to each other and electrical signals are generated on the basis of the variation of light passing through these gaps, or the interferential scanning principle, which exploits the diffraction and interference of light on a fine graduation to produce signals used to measure displacement [2].

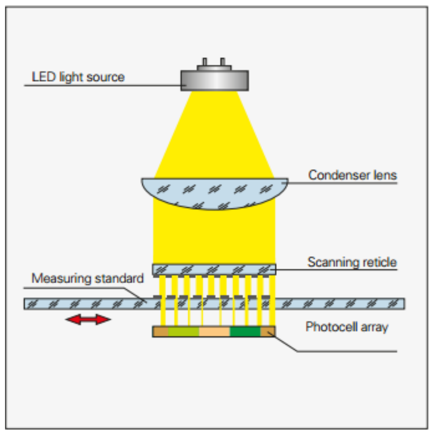

Figure A-2. Imaging Principle Scanning Method [2]

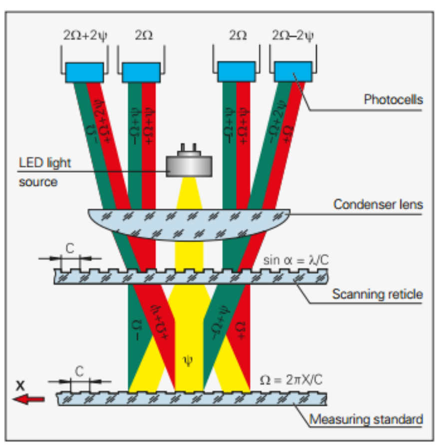

Figure A-3. Interferential Scanning Principle [2]

Digital Image Correlation

Digital Image Correlation (DIC) is an optical method that employs pattern matching and image registration techniques for accurate two- and three-dimensional measurements of changes in the shape of an item. It is thus a non-contact and non-destructive method. It can be used to measure change of shape of an object, measuring the object's deformation, displacement, and change in strain [3].

In performing DIC, the surface of the specimen or area to be tested is sprayed or marked with a dot pattern, and targets are used to calibrate the system. A digital camera, or two if the specimen needs to be analyzed in three-dimensions, then takes high resolution photographs of the specimen surface at the beginning of the test, before any stress or deformation has occurred, as well as at regular intervals during the testing. The specimen's changes over time, as well as total displacements or strains, can then be studied in detail [4].

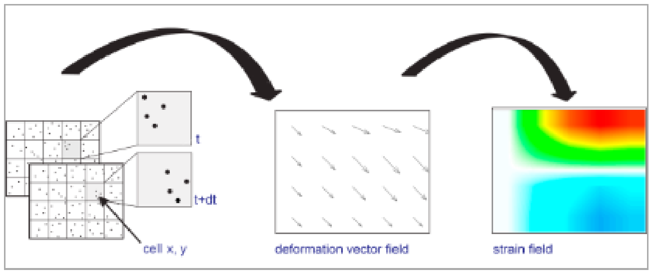

Figure A-4. Digital Image Correlation Process [5]

DIC's Nuclear Applications

Digital Image Correlation has been used in relation to nuclear power plants before, often being used for structural integrity tests of a nuclear power plant's containment building. However, the testing only called for increased pressures, not temperatures, and the walls of the containment were able to be imaged and analyzed easily.

When considering a material for future power plant operations, it would be necessary to test a specimen of that material in plant like conditions or a plant itself. This presents new challenges of how to mark and decipher images of the sample when red hot or in close quarters that make for a sample moving out of range.

These considerations could be addressed prior to testing through their own testing, such as imposing a grid on the specimen that could be seen at the test temperature, or even drilling small markings into the sample.

Multiple cameras could also be used to ensure the specimen is always in view, or the specimen should be centered in the camera, so that the elongating portion is centered and strain can still be measured. An additional strain gauge could be bonded to the specimen to provide a basis for comparing the DIC results to the measured strains, perhaps a bonded resistance gauge as it is small, inexpensive, and only moderately impacted by temperature changes, and could thus provide a good basis for comparison to the DIC measured strains [6].

Tests have also been run where DIC has been used on in-cell mechanical tests through the thick, leaded-glass shield windows with a correction methodology implemented [7]. If feasible, this could also help ensure the specimen is able to be imaged throughout the entire testing procedure.

Direct Current Potential Drop Method (DC/PD)

Aggressive environments at high temperatures can significantly affect the creep crack growth behavior of a material. The direct current potential drop method is a method that helps scientists understand these effects by covering the determination of creep crack initiation and creep crack growth in metals at elevated temperatures using pre-cracked specimens subjected to static or quasi-static loading conditions [8].

The potential drop method entails passing a constant current (maintained constant by external means) through a cracked test specimen and measuring the change in electrical potential across the crack as it propagates. With increasing crack depth, the uncracked cross-section of the test piece decreases and its electrical resistance increases, raising the potential difference between two points spanning the crack. By monitoring this increase and comparing it with a reference value measured at the start of the test, the crack depth or the crack depth-to-specimen width ratio can be determined by means of appropriate calibration curves [9].

The crack growth derived in this manner is identified as a material property which can be used in modeling and life assessment methods of the material and its applications [8].

This technique is also attractive for many other reasons. The potential drop method can be used for any geometry and continuous monitoring of the crack depth can be achieved. Only simple instrumentation (and thus low costs) is required, and it is stable and suitable for automation (continuous output). This technique also allows for very small increments of length to be detected and non-uniform crack development to be monitored. Also visual access is not required, making the technique suitable for sealed environments. It is also a physically robust method and could potentially provide in situ crack growth monitoring of real power-plant components [9].

However, drawbacks of the technique include the fact that a calibration curve is required for each geometry, and underestimation of the crack depth can potentially occur should crack faces come into contact creating a short circuit situation [9].

Fabry-Perot

A small cavity is formed at the end of a fiber with a reflective surfaces where multiple reflections occur. The round-trip propagation-phase-shift, φ, in the cavity is:

$$\varphi = 4\pi nL/\lambda$$

where n is the index of refraction of the cavity, L is the spacing between the reflective surfaces, and λ is the free space optical wavelength. The reflective surface opposite the fiber flexes in response to a pressure differential between the cavity and the environment. The strong advantage of the interferometric measurement (and other interferometric) compared to intensity-modulated sensors is the measurement of phase difference, making the measurement insensitive to absolute measurements. Phase measurement further ensures successful measurement even with potential darkening effects.

Hollow core photonic crystal based spectrometer

Hollow core photonic crystal fibers use photonic bandgap effect provided by the periodic microstructure air holes in the cladding to guide light into the core. The composition of gas in the hole will change the absorption of light guided into the fiber following Beer-Lambert law (absorption spectroscopy). Through measuring the intensity of the wavelengths collected at the receiving end, the exact composition of gas is determined. Another way to determine the composition of the gases is through etching gratings (Fiber Bragg Grating or Long period grating) on these fibers and look at the reflected/transmitted light. The wavelength shift (resonance shift) will determine the composition of the gas. The fiber can also work as a pressure sensor as the wavelength shift is also dependent on the pressure.

Fiber optic based temperature sensing (Nirmala Kandadai, Harish Subbaraman, Lan Li).

Fiber optics sensors have a significant advantage over conventional temperature sensors. They are robust, compact, efficient, immune to electromagnetic interference, passive and resistant to electromagnetic interferences. Two classes of fiber gratings can be used as sensors for temperature: One is fiber Bragg grating, such gratings works on Bragg condition, where the reflected light from the grating is dependent on the refractive index. The fiber Bragg grating works as a temperature sensor due to its thermo optic effect, where change in temperature changes the refractive index and hence changes the wavelength of the reflected light.

Second is a long period grating, where the light transmitted through the fiber is dependent on the refractive index of core and cladding. In this case, the fiber couples the fundamental mode with modes propagating in the cladding. The transmission spectrum has a series of attenuation band centered at resonant wavelength. The resonant wavelength is dependent on the refractive index of the core and cladding. Similar to fiber Bragg grating, the long period grating also experience thermos optic effect leading to a shift in the attenuation band.

Early research in using fibers as a sensor failed as they were known to darken with exposed radiation. Recent advances in fiber optic technology have shown that fibers can be generated with high tolerance to gamma radiation and have low to moderate radiation induced attenuation. Doped fibers, Sapphire fiber, hollow core photonic crystal fiber, or FBG based on photonic crystal fiber have shown promises [.

Manufacturing the fiber grating has also undergone tremendous improvement. Ultrafast laser etched fiber Bragg gratings have shown higher tolerance to radiation than conventional gratings. Femtosecond laser etching uses threshold dependent multiphoton ionization process to inscribe gratings. These type of gratings are known as Type II gratings or damage gratings. A primary advantage of such etching is that it is not limited to Silica fibers but can be used for any fiber material. For example femtosecond IR laser etched grating on fluorine doped fiber showed very little loss of spectral quality even with 100 kGy gamma radiation. In this area we also explore additive manufacturing which allow cheap, low cost sensors.

Optical Fiber-Based Neutron Detection (Nirmala Kandadai, Harish Subbaraman, Kurtis Cantley)

Fiber optics make an attractive sensor as they are non-contact, easily deployable, robust and insensitive to electromagnetic radiation. Recent research has used fiber optics to detect neutron flux by combining them with scintillators. The scintillators generate light which is transmitted through the fiber, and a photomultiplier is used to detect the signal away from the radiation zone. Three combinations of fiber optics with scintillators are used – fiber with its tip coated with a scintillator, fiber optic material with a scintillator dopant or a combination of both.

Nano-structured Materials for Solid-State Neutron Detection (Liz Godwin, David Estrada, Kurtis Cantley)

Detectable byproducts of neutron interactions will create electron-hole pairs inside the intrinsic region of diodes, measured as a current in the external circuit. Even in configurations requiring multiple converter layers or materials, clever design of physical device structures (especially those potentially compatible with 3D printing) will enable maximized detection efficiency.

Aerosol Jet Printing

Aerosol Jet printing technology effectively produces 2D/3D printed sensors and electronics. It uses an additive manufacturing process that directly prints conductive, dielectric, semiconductor and other sensor materials onto a variety of 2D or 3D plastic, ceramic, and metallic substrates. Aerosol jet based sensor printing provides several significant advantages over current approaches:

The AJP printing allows 3D conformal sensor printer directly onto the fuel component of any 3D geometry. This allows nonintrusive sensor implementations with intimate thermal and mechanical contacts, which allows the thermal conductivity measurement with high accuracy.

The AJP printing enables sensor printing of superior spatial resolution (~10 μm). The microscale printing resolution not only allows printing sensor arrays with high spatial resolutions, but also result in high temporal resolutions due to the small sensor thermal masses. The AJP printing has been widely used in printing sensors for extreme conditions, such as aerospace and turbine engines. This in-pile initiative can leverage rich knowledge generated from these relevant fields, and will produce new knowledge on printed sensor performances in nuclear energy environment.