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Thermal Properties Capability Development Workshop Summary to Support the Implementation Plan for PIE Thermal Conductivity Measurements

Lori Braase, Cynthia Papesch

April 2015

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Fuel Cycle Research & Development

April 30, 2015

Prepared for
U.S. Department of Energy
Advanced Fuels Campaign

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EXECUTIVE SUMMARY

Nuclear technologies require in depth analysis of material performance under extreme conditions to develop new composite materials and nuclear fuels to meet the system requirements. Thermal properties of nuclear fuels and materials are key attributes to understand and predict the performance of the fuels and materials in the reactor system. Therefore, measurement techniques are required to analyze the materials at various length scales, from bulk properties to microscale.

Measuring thermal properties on irradiated fuels and materials adds difficult layers of complexity, including shielding; sample preparation, transfer, and handling; instrument capability; technique; and analysis. This can be achieved either by developing new instruments/techniques or by modifying the existing ones to improve reliability and operability under irradiated conditions.

The Department of Energy (DOE)-Office of Nuclear Energy (NE), Idaho National Laboratory (INL), and associated nuclear fuels programs have invested heavily over the years in infrastructure and capability development. With the current domestic and international need to develop Accident Tolerant Fuels (ATF), increasing importance is being placed on understanding fuel performance in irradiated conditions and on the need to model and validate that performance to reduce uncertainty and licensing timeframes.

INL's Thermal Properties Capability Development Workshop was organized to identify the capability needed by the various nuclear programs and list the opportunities to meet those needs. In addition, by the end of fiscal year 2015, the decision will be made on the initial thermal properties instruments to populate the shielded cell in the Irradiated Materials Characterization Laboratory (IMCL).

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FIGURES

TABLES

ACRONYMS

AC	alternating current
ATF	Accident Tolerant Fuels
DC	direct current
DCS	differential scanning calorimetry
DOE	U.S. Department of Energy
DOE-NE	DOE Office of Nuclear Energy
EBSD	electron backscatter diffraction
FFG	Fresh Fuels Glovebox
FIB	focused ion beam
FY	fiscal year
HFEF	Hot Fuels Examination Facility
IMCL	Irradiated Materials Characterization Laboratory
INL	Idaho National Laboratory
ISU	Idaho State University
ITU	Institute for Transuranic Elements
MOOSE	Multiphysics Object-Oriented Simulation Environment
NEAMS	Nuclear Energy Advanced Modeling and Simulation
NGT	Nominal Group Technique
PPMS	Physical Properties Measurement Systems
RISE	Research and Innovation in Science and Engineering (complex)
SEM	scanning electron microscope
TCM	thermal conductivity module

THERMAL PROPERTIES CAPABILITY DEVELOPMENT WORKSHOP

1. Introduction

Nuclear technologies require in depth analysis of material performance under extreme conditions to develop new composite materials and nuclear fuels to meet the system requirements. Thermal properties of nuclear fuels and materials are key attributes to understand and predict the performance of the fuels and materials in the reactor system. Therefore, measurement techniques are required to analyze the materials at various length scales, from bulk properties to microscale.

Measuring thermal properties on irradiated fuels and materials adds difficult layers of complexity, including shielding; sample preparation, transfer, and handling; instrument capability; technique; and analysis. This can be achieved either by developing new instruments/techniques or by modifying the existing ones to improve reliability and operability under irradiated conditions.

Typical thermal properties include thermal conductivity, thermal diffusivity, and specific heat capacity, and are used to define a material's ability to store and transfer heat. These properties are essential for nuclear fuel/materials design, performance in the reactor, and predicting/modeling fuel behavior. Thermophysical property measurements also reflect important information about material composition, purity, and structure, as well as secondary performance characteristics such as tolerance to thermal shock. Materials selection decisions for components that are exposed to elevated temperature changes and/or thermal gradients require an understanding of the thermal responses of fuels and materials.

The Department of Energy (DOE)-Office of Nuclear Energy (NE), Idaho National Laboratory (INL), and associated nuclear fuels programs have invested heavily over the years in infrastructure and capability development. With the current domestic and international need to develop Accident Tolerant Fuels (ATF), increasing importance is being placed on understanding fuel performance in irradiated conditions and on the need to model and validate that performance to reduce uncertainty and licensing timeframes.

The Fuel Cycle Research and Development (FCRD) program has been tasked with supporting development of post irradiation characterization of thermal properties on relevant nuclear fuels and structural materials. As part of the work being conducted by the FCRD program the Thermal Properties Capability Development Workshop was organized to identify the capability needed by the various nuclear programs and list the opportunities to meet those needs. In addition, by the end of fiscal year (FY) 2015, the decision will be made on the initial thermal properties instruments to populate the shielded cell in the Irradiated Materials Characterization Laboratory (IMCL).

This document summarizes the output from the Thermal Properties Capability Development Workshop, which will be used to inform future planning and funding decisions at INL and within the various nuclear programs. The meeting agenda and attendees are included in the appendices for reference.

2. Workshop Overview

The objectives for the workshop were to develop the strategy for thermal properties capability development at INL and to identify the potential suite of thermal property measurement equipment for IMCL. Several subject matter experts were asked to provide presentations to set the background and inspire thought on the workshop content. These presentations were followed by a Nominal Group Technique to collect ideas and input from the workshop participants.

Jon Carmack, Advanced Fuels Campaign National Technical Director, discussed the drivers for thermal properties capability development at INL. The combination of the short timeframe to develop ATF, create the codes and models to support licensing, and meet the goal for improved safety in accident conditions are strong drives to meet program goals for 2022. Other nuclear programs have critical drivers as well, such as transient testing of fuels, including ATF, first-of-a-kind fuel development to support industry, improved reactor performance, and long-term storage of used nuclear fuel.

Participants were encouraged to think broadly about their needs for thermal property measurements on a wide range of applicable fuels and materials. There are many philosophies and approaches to thermal property measurements, and the goal is to bring all those ideas together to decide how to *push the science forward*.

Idaho National Laboratory – Pushing Science Forward ...

David Hurley, Materials Science, presented "Thermal Property Measurements – Accuracy and Reproducibility." This included an overview of two categories of measurement techniques, the direct current (DC) method and alternating current (AC) method, which is a little more complicated. Large sample accuracy is not an issue, but smaller samples become more difficult to accurately measure. (See Appendix C)

Cynthia Papesch, Materials Characterization, presented "Current Thermal Properties Capability at INL." This was an overview of facilities and specific thermal properties measurement capability that is available at INL. A discussion followed about the important parameters unique to irradiated fuel measurements, such as specialized techniques and glovebox atmosphere. (See Appendix D)

The "Modeling and Simulation Perspective on Thermal Transport Measurements," was presented by Mike Tonks, Nuclear Energy Advanced Modeling and Simulation (NEAMS). It is important to understand thermal transport, which is when heat is conducted through convection, radiation, and conduction. Understanding thermal conductivity is needed to understand thermal transport. (See Appendix E)

Progress has been made on mechanistic understanding of what is occurring, but the validation step is missing. Measurements are needed on local thermal conductivity of all phases to validate thermal conductivity models. This would provide a validated answer and confidence on predictions of fuel behavior. Thermal conductivity measurements would be useful in validating our modeling and simulation. Data is needed to understand and model thermal transport, but that won't happen without detailed understanding of the microstructure.

3. Nominal Group Technique – Nuclear Program Needs for Thermal Property Measurements

Nominal Group Technique (NGT) was used to gather thermal property data "needs" from the participants. NGT is a facilitated group process used to collect data quickly and to encourage equal participation. This technique improves effectiveness of decision-making groups by asking individuals to write down their ideas silently and independently prior to a group discussion. Then, by round-robin polling, ideas are collected, duplicate ideas are eliminated, and similar ideas are grouped in preparation for ranking. The typical benefits of NGT are unique ideas, balanced participation between group members, increased feelings of accomplishment, and greater satisfaction with idea quality and group efficiency. The ideas were captured on flip charts and similar ideas were grouped together based on team consensus. Each member was given 5 dots, numbered from 1-5. They were instructed to place the #1 dot next to their #1 "most important need," and so forth. Results of this activity are shown in Table 1. The items in orange were further discussed by Team 1; items in green were discussed by Team 2.

Table 1. NGT Consensus Voting Results

NGT Question: What thermal property measurements or results are needed by the programs?			
Rank	**Measurements or Results Needed**	**# of Votes**	**Total Score**
1	Thermal conductivity or diffusivity of individual microstructural features – both fresh and irradiated • Defects in isolation • Individual features • Point – line – planar – volume • Multiple phases	13	50
2	Bulk thermal properties on both fresh and irradiated fuels	12	48
3	Good characterization of samples – full amount of properties on fresh and irradiated	8	25
4	Thermal conductivity across whole fuel system from pellet to cladding to gap.	4	15
5	Influence of multiple defects in isolation versus cooperative effects • Synthesis and characterization of samples with multiple defects	3	14
6	Systematic data; thermal conductivity on fuels. More data points and microstructural variation	3	12
7	Single crystal measurements as a function of stoichiometry	5	10
8	In-pile measurements	5	9
9	Data needs to have uncertainty quantification	4	9
10	Bulk techniques to measure smaller samples (1mm)	3	9
11	Comparison of different measurement methods	4	9
12	NDE 3-D characterization. Provide characterization of stoichiometry; secondary phases	3	6
13	Wide temperature range – very low to very high	2	6
14	Thermal property measurements on a variety of burnup or irradiation condition. General purpose tool to use 12 months after irradiation on uranium compounds that vary from insulated to spectrum of fuels. Metals to ceramic.	1	5
15	TC/TP irradiated steels (cladding / structural materials)	1	4
16	Different interaction of fuels. Thermal conductivity in extreme conditions. Specific heat – high pressure – magnetic field, etc.	1	4
17	Impurity effects	1	3
18	Ability to measure irregular shapes	1	3
19	High temperature; radial profile and large temperature gradients	1	2
20	Measure at microstructure level to develop and inform; multiscale simulation	1	2
21	High temperature drop calorimetry for specific heat	1	1
22	Atomistic measurements to develop sub-microstructure material. Investigating magnetic influence as well as other things such as oxygen, atom defects – across grain boundaries	1	1
23	Data collected needs to be applicable to NRC licensing	1	1
24	Measure thermal conductivity not only at the surface of material but also at distance	1	1
25	Effect of fission products in general. Metallic participate versus other things that form within the fuel.	1	1
26	Data relevant to fuel performance calculations	1	1
27	Interface resistance, such as grain boundaries – interface – cold be all scales	0	0
28	Measurements on a variety of materials; full spectrum of fuel types.	0	0
29	Thermal conductivity of liquid materials	0	0
30	Fresh fuel – magnetic spin – fundamental condition mechanism (without defects)	0	0
31	Correlate fission gas release with TC, enthalpy, etc.	0	0

NGT Question: What thermal property measurements or results are needed by the programs?			
Rank	**Measurements or Results Needed**	**# of Votes**	**Total Score**
32	Emissivity	0	0
33	Radial measurements – cross-section	0	0

4. Breakout Session – Detailed Discussion of Thermal Property Needs

Breakout sessions were conducted to further define the highest ranking thermal property needs. A matrix was used to guide the discussion and collect additional data. The matrix questions were as follows:

- What measurements are needed? What do you need to know?
- How can the need be met? Combinations? Instruments? Processes?
- Does it (the capability) exist? Need modification? Need development?
- Is it (the capability) available? Less than two year? Two to five years? Five to ten years?
- What is the benefit of the capability?
- What are the barriers to success?

4.1 Team 1

The thermal property needs discussed by Team 1 are provided in this section, and include the following:

- Bulk thermal properties on both fresh and irradiated fuels
- Thermal conductivity across the entire fuel system
- Comparison of different measurement methods.

4.1.1 Bulk Thermal Properties on both Fresh and Irradiated Fuels

What measurements are needed? Thermal conductivity, specific heat, thermal expansion, density, and thermal diffusivity on composite fuel material are the properties that were identified as necessary to fulfill the programmatic requirements – multiphase, engineering properties, effective values averaged over mm length scale (vs. µm). This is for all operating and accident conditions as well as all fuel types, including metal, ceramic, high-conductivity, low-conductivity, etc. The focus of this discussion was on the fuel pellet, and not cladding materials. However, the external corrosion scale and cladding may still need to be addressed.

How can the need be met (capability)? The following list addresses how the INL can meet the needs to measure the properties identified above. The ability to measure smaller sample sizes on a laser flash analyzer is critical to moving on with thermal properties capability development.

- Fresh Fuels Glovebox – contains instrumentation capable of measuring thermal conductivity from room temperatures to 1650°C
- Calorimetry (i.e., differential scanning calorimetry [DSC]) to measure specific heat
- Push rod dilatometer to measure thermal expansion and density as a function of temperature
- Pulse laser flash analyzer to measure thermal diffusivity/conductivity/)
- Further development of the Physical Properties Measurement System (PPMS) could be useful to measure thermal conductivity on small samples (approximately 1mm) at room temperature or subambient temperatures
- AC techniques – to measure thermal diffusivity

- DC techniques – These were discussed, but since they are not used much as industry standards and show a lot of heat loss during use, it was determined that INL will not investigate these methods further.

Does the capability exist, need modification, or need to be developed? When will it be available? The capability for measuring thermal properties exists today for fresh fuel in the Fresh Fuel Glovebox. However, the current instruments may not go to high enough temperature for all fuels nor could they cover all sample size ranges that have been identified. Success demands proper atmospheric control for all fuel types.

The capability to measure bulk thermal properties on irradiated fuel does not exist and should be developed within approximately two years. Irradiated work with this suite of instrumentation would require a determination of how to operate them remotely in a high-level radiation area. Sample sizes would need to be identified for typical types of irradiated fuel. Unique sample preparation on these irradiated fuel samples will need development as this is not a trivial activity.

What is the benefit of the capability? A thermal property measurement capability provides engineering scale properties that are directly applicable to calculations and fuel design for both fresh and irradiated fuels and materials. It also provides data needed for validating the lower-length scale model development (see Team 2 topics) and separate effects validation at engineering scale. With the ability to measure smaller sample sizes, samples can be measured out of the reactor sooner with less exposure, less sample preparation, and reduced shielding requirements.

What are the barriers to success? Below is a listing of barriers to successful implementation of a thermal properties measurement capability on irradiated fuels and materials.

- Implementing equipment remotely in a high-radiation environment. Proposed hot cell space is available in the IMCL. Most of these types of instrumentation have very delicate components, which will add to this challenge.
- Effects of sample radioactivity on instrument
- Sample preparation methods need development. An example would be that it is necessary to prepare samples that preserve the cracks that develop during irradiation for the actual measurement.
- Available furnace temperatures
- Available reference materials for specific heat are limited. This is an area where the programs could work on developing a new reference material.
- Established techniques require larger samples that are easy to get with fresh fuel; however, smaller sample size and preparation in a hot cell will be more difficult.
- Currently samples are cut axially from an irradiated fuel rod. This direction does not follow the natural heat flow direction, therefore making any data collected not representative of the true behavior of the fuel. Preparing a sample in the longitudinal direction is significantly more challenging.

4.1.2 Thermal Conductivity across the Whole Fuel System

What measurements are needed? The ability is needed to measure thermal conductivity across the fuel system, from the pellet through the gap and the cladding is crucial in understanding the performance of the fuel assembly. Cladding is generally more understood within this system, with the exception of the surface condition of the cladding. Collecting data on the gap is also very challenging since there are lots of uncertainties in gap conductance models (e.g., roughness effects and jump distance and contact pressure).

How can the need be met (capability)? Off-the-shelf instruments for this type of thermal conductivity measurement do not currently exist. It is a unique experimental setup, which could be set up quickly in a laboratory setting with the right approach. One idea would be to develop a measurement method for an entire cross section.

Does the capability exist, need modification, or need to be developed? When will it be available? The capability does not currently exist, but with priority and funding, it could be developed within two years. Better experimental measurement of parameters in the models is needed. Initial design of this experimental set up does not have to work with nuclear materials, so building and operating an experiment should be much easier.

What is the benefit of the capability? The measurement capability removes uncertainty from predictions (the greatest uncertainty is associated with the fuel/cladding gap).

What are the barriers to success? The equipment to measure thermal conductivity across this system has to be invented. It is important to note that at high temperatures, emissivity becomes a parameter that will be needed as well.

4.1.3 Comparison of Different Measurement Methods

What measurements are needed? Comparison of different methods can be used to measure the same sample and, by doing so, increase uncertainty quantification in thermal property and microstructure measurements. An example of this comparison would be to correlate physical properties and microstructural characterization on the same sample.

How can the need be met (capability)? Standard materials, although less desirable, or samples fabricated from surrogates could be used for comparison purposes. Using different methods to measure the same samples at the same location or length scale and then comparing the results will add to the understanding of the relationship between macro and microscale properties.

Does the capability exist, need modification, or need to be developed? When will it be available? Developing the comparison method should not be very complicated, but it has not been done before. With funding and resources, it could be developed within two years.

What is the benefit of the capability? The measurement capability will quantify uncertainty within results reported from experiments. This type of measurement comparison is a good way to validate new measurement techniques.

What are the barriers to success? This measurement technique requires many different instruments, some of which are available at universities and some of which are at INL. Therefore it is possible that samples would have to be transferred out of INL during this capability development. Fabricating the sample/surrogate to have a reproducible microstructure and different length scales could be difficult.

4.2 Team 2

The thermal property needs discussed by Team 2 are provided in the bullets below. Clarification was made between engineering, science, and validation. Engineering is focused on understanding a specific fuel composition and irradiation condition. Science identifies structures relevant to the sample. Validation is for fuel performance modeling. Identified thermal property needs are as follows:

- Single crystal measurements as a function of stoichiometry
- Influence of multiple defects in isolation versus cooperative effects; synthesis and characterization of samples with multiple defects
- Thermal conductivity or diffusivity of individual microstructural features in both fresh and irradiated materials:
  - Defects in isolation
  - Individual features
  - Point – line – planar – volume
  - Multiple phases
- Bulk techniques to measure smaller samples (<1mm).

4.2.1 Single Crystal Measurements as a Function of Stoichiometry

What measurements are needed? Measuring the complete conductivity tensor of single crystal urania as a function of off-stoichiometry would be beneficial. There is no source and few samples because it is hard to grow crystals. Urania is plausible on a scale of 1mm x 5 x 5. Stoichiometry is needed on poly crystals.

How can the need be met (capability)? Experiments could be done on legacy materials. Bulk measurements can be done on single crystals. The TCM can used to verify the recently proposed models that suggest thermal transport in UO2 is anisotropic. The ability to control stoichiometry is needed. Single crystals could be measured with a PPMS or thermal flash.

Does the capability exist, need modification, or need to be developed? When will it be available? Materials are available for low-temperature data, but materials are missing for high-temperature data. Measurements could be made on unirradiated, low-temperature materials. Some experiments will be done in the next 6 months.

What is the benefit of the capability? The capability gathers data on the basic mechanisms of heat transport of UO_2 and starts to look at defects. A single crystal baseline is needed.

What are the barriers to success? Programs are not funding these fundamental measurements on UO_2. There is a risk of synthesis. Samples may be available through the Research and Innovation in Science and Engineering (RISE) facility at Idaho State University (ISU).

4.2.2 Influence of Multiple Defects in Isolation versus Cooperative Effects; Synthesis and Characterization of Samples with Multiple Defects

What measurements are needed? Segregation of point defects at the grain boundary is needed.

How can the need be met (capability)? A modified thermal conductivity module (TCM) should be considered.

Does the capability exist, need modification, or need to be developed? When will it be available? Not identified.

What is the benefit of the capability? Not identified.

What are the barriers to success? Not identified.

4.2.3 Influence of Isolated Microstructural Features on Thermal Transport

What measurements are needed? The ability to measure influence of microstructure on thermal transport will require the development of new fabrication and characterization capability. These measurements are complicated by the need to produce samples with tailored microstructure in which thermal transport is determined solely by a single feature type. It is both difficult to fabricate distinct and quantifiable microstructural features and hard to measure thermal transport effects on such features. Current work is focusing on the measurement techniques. The following list provides further detail into the challenges associated with development of this measurement capability.

- Measurement of point defects in unirradiated fuel. Concentrations and secondary characterizations (depending upon the defect) could be measured with a Laser Flash, PPM, or TCM in the next two years. This would provide the ability to measure the change in thermal conductivity as a result of the presence of point defects. Dispersed point defects are the most difficult to characterize and yet have the most impact on thermal transport.
- Planar or line defects; volumetric measurement. Some the defects can only be made with an ion beam, which restricts the ability to measure transport. Measuring thermal transport in ion irradiated samples requires characterization techniques with high spatial resolution such as the TCM. Microscopy could be used to look at the concentration of these defects. Fabricating samples with planar defects can be done today. Engineering samples that are irradiated would need to go to IMCL. The capability can be made available in a two to five year timeframe.
- Fabrication Science. This can be done with ion irradiation, fabrication with isotopes that decay into fission gas, or a TCM down to 10 microns. The benefit is controlled microstructures in non-radioactive samples.
- Integral irradiation tests. Spatially resolved characterization techniques, such as the TCM, could be used to build a 3D map of thermal properties with micron resolution. For this approach a plasma focused ion beam (FIB) is an option to peel away successive layers to assist in building a 3D property mapping of the sample of interest. The program should consider purchasing a plasma FIB in the five to 10 year time frame. Ultimately, we should consider integrating a FIB, electron backscatter diffraction (EBSD), and TCM to create a super instrument. The benefit from these options is a layered 3-D approach to thermal conductivity with characterization. The ability to benchmark Multiphysics Object-Oriented Simulation Environment (MOOSE) calculations for integral tests is also an advantage.

How can the need be met (capability)? (See bulleted items above.)

Does the capability exist, need modification, or need to be developed? When will it be available? (See bulleted items above.)

What is the benefit of the capability? (See bulleted items above.)

What are the barriers to success? (See bulleted items above.)

4.2.4 Bulk Techniques to Measure Smaller Samples (<1mm)

What measurements are needed? Bulk measurements on small samples are needed. The majority of currently available off-the-shelf instruments require a large variety of sample dimensions most of which are larger than the dimensions required for fabricating samples in a test reactor. Therefore, the fuels programs are limited in the sizes of samples available on post-irradiated materials.

How can the need be met (capability)? The need can be met by modifying a laser flash to have a smaller spot size. With the PPMS, the heat capacity is much less than 1mm. For conductivity measurements, it would be two times larger than 2mm cubed. Literature from PPMS states that for the thermal conductivity range of interest for current and ATF fuel, the sample must be 8mm thick.

Does the capability exist, need modification, or need to be developed? When will it be available? The most straight forward approach would be to increase the field of view of the TCM or decrease the field of view of pulsed laser methods.

What is the benefit of the capability? The ability to measure spent fuel fragments or other small regions of interest would become possible and have the possibility of a simpler sample preparation technique.

What are the barriers to success? None identified.

5. IMCL Overview and Requirements/Constraints for TP Capability – Collin Knight

The IMCL at INL has a designated shielded space for a thermal property measurement capability. The current cell is 10' x 10' and is shown on the IMCL floor map in Figure 1. If additional space is needed, it could be expanded 1-2 feet or relocated to a different area (i.e., the 10' x 20' "Future Prototyping Area." Is available). The thermal properties cell will likely be fully enclosed. It will have limited remote handing capability. If something breaks, the shield door would be opened for repairs.

For comparison, the Fresh Fuels Glovebox (FFG) is 35 feet long, so the measurement capability wouldn't fit in the prototyping area because of the 20' wide limitation. Therefore, the idea of replicating the FFG to meet irradiated property measurement needs is not the ideal approach. There is about $3.5M available to design, fabricate, and deliver a 10' x 10' thermal properties hot cell. It is important that the requirements are identified along with expected materials and output data so the cell can be designed appropriately.

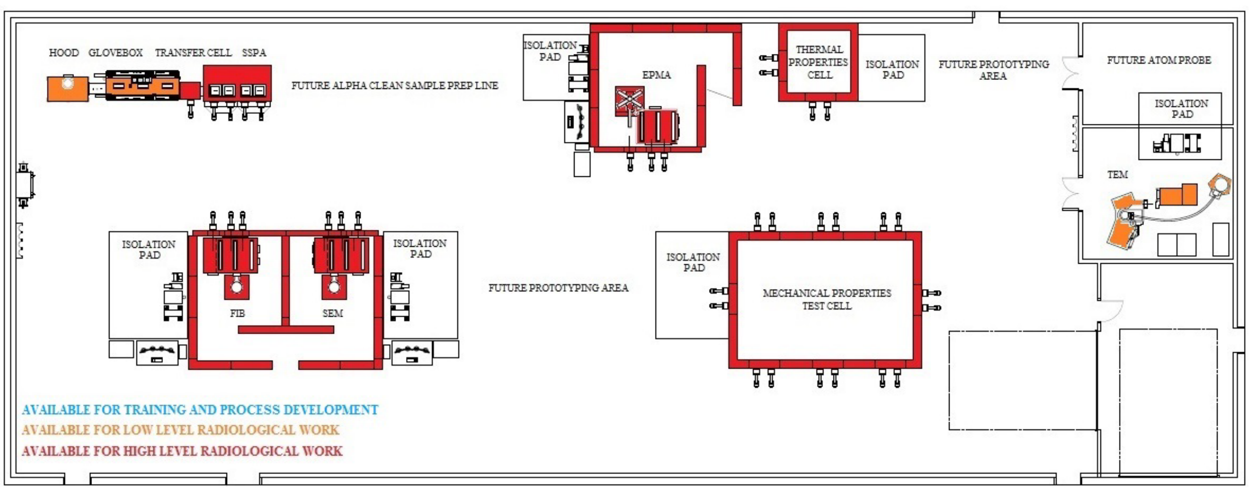

Figure 1. IMCL Floor Plan in 2018.

6. IMCL Closing Discussion

By the end of FY 2017, thermal property measurement capability will be installed in IMCL. The group discussed the types of equipment and constraints in IMCL.

- This is not a one-to-one duplication of the Fresh Fuels Glovebox.

- The sample size will not be a fuel pin. It will be larger than a typical sample and will require a very specific sample preparation
- Initial measurements for ATF in the 2017-2018 timeframe should use a phased approach. Initial measurements would include bulk and other typical thermal properties capability development strategy and should stay flexible to adapt to new techniques and instruments. The flexible philosophy for thermal properties will be able to meet multiple needs. That is what will make it usable and desirable.
  - Laser flash – Off-the-shelf unit has a fixed size laser beam.
  - Laser flash – INL-built with a smaller sized laser beam – not off-the shelf.
- An advantage of laser heating is that higher temperatures can be achieved when you can heat a small sample effectively.
- Inert atmosphere control is exceptionally critical.
- Sample preparation will be different for different equipment and measurements.
  - TCM vs Laser Flash
  - Need a sputter coater capability
  - Initially, most sample preparation will start in the containment box at the Hot Fuels Examination Facility (HFEF). This step must be done right the first time, so it does not have to go back to HFEF.
  - Some materials are better prepared in air (Note: difference between inert atmosphere and high purity atmosphere).
  - A pass through Argon box is needed for preparation of samples that are oxygen sensitive.

IMCL Phased Thermal Properties Implementation (discussion)

- Phase 1 – Initial complement of the suite of instruments needed to measure thermal properties of ATF (Initially, the suite of equipment will be used to meet ATF requirements).
  - Institute for Transuranic Elements (ITU) Laser Flash is variable and easy to customize. (Note: laser flash does not directly measure thermal conductivity).
  - TCM
  - DSC and dilatometer.
- Phase 2 – Metal fuels at higher temperatures (after completion of initial ATF samples).
- Phase 3 – Procedural
  - How to use existing equipment differently
  - Suite of instruments in one place
  - How to look at something under the scanning electron microscope (SEM) so it can be taken to TCM
  - Explain in a "measurement plan" the suite of instruments and capability in IMCL and how to get the measurements needed.
  - Make equipment and space reconfigurable for modeling and simulation in Phase 3.

7. Summary

The objectives for the workshop were to develop the strategy for thermal properties capability development at INL and to identify the potential suite of thermal property measurement equipment for IMCL. The ideas generated by the attendees will be used to develop an implementation plan for thermal property capability development at INL. They will also be valuable information for future planning and funding decisions within the various nuclear programs.

Appendix A: Agenda

Thermal Properties Capability Development Workshop

Idaho National Laboratory – Energy Innovation Laboratory (EIL) April 14, 2015

AGENDA

Objectives:

- Develop the strategy for thermal properties capability development at INL.
- Identify the potential suite of thermal property measurement equipment for IMCL.

8:30	Agenda/Guidelines/Introductions	Lori Braase
8:50	Welcome/Expectations	Jon Carmack
9:00	Thermal Property Measurements – Accuracy and Reproducibility	David Hurley
9:30	Current Thermal Properties Capability at INL	Cindi Papesch
10:00	Modeling and Simulation Perspective on TP Measurement	Mike Tonks
10:30	***Break***	
10:45	Nominal Group Technique: Identify/prioritize program needs for TP measurements. What measurements or results are needed? (Function – not equipment)	Darcie Martinson
11:45	***Lunch – On your own***	
1:00	For each program need, HOW can it be measured? (Equipment – new or modified?) WHAT capability is missing (gaps)? HOW do we fill the gaps? WHEN is it needed? WHERE (location)? WHAT are the barriers to success?	Breakout Groups (2 to 3)
2:45	***Break***	
3:00	Breakout Reports	
3:45	IMCL Overview and requirements/constraints for TP capability	Collin Knight
4:30	Identify initial compliment of TP measurement instruments for IMCL	All
5:00	Path Forward / Actions / Adjourn	Lori Braase

Appendix B: Attendees

First Name	Last Name	Phone	Email	Role	Org
David	Bai	208-526-2496	Xianming.bai@inl.gov	Fuels Modeling & Simulation	INL
Lori	Braase	208-526-7763	lori.braase@inl.gov	Systems Engineer	INL
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Andy	Nelson	505-667-1268	atnelson@lanl.gov	AFC Ceramic Fuels	LANL
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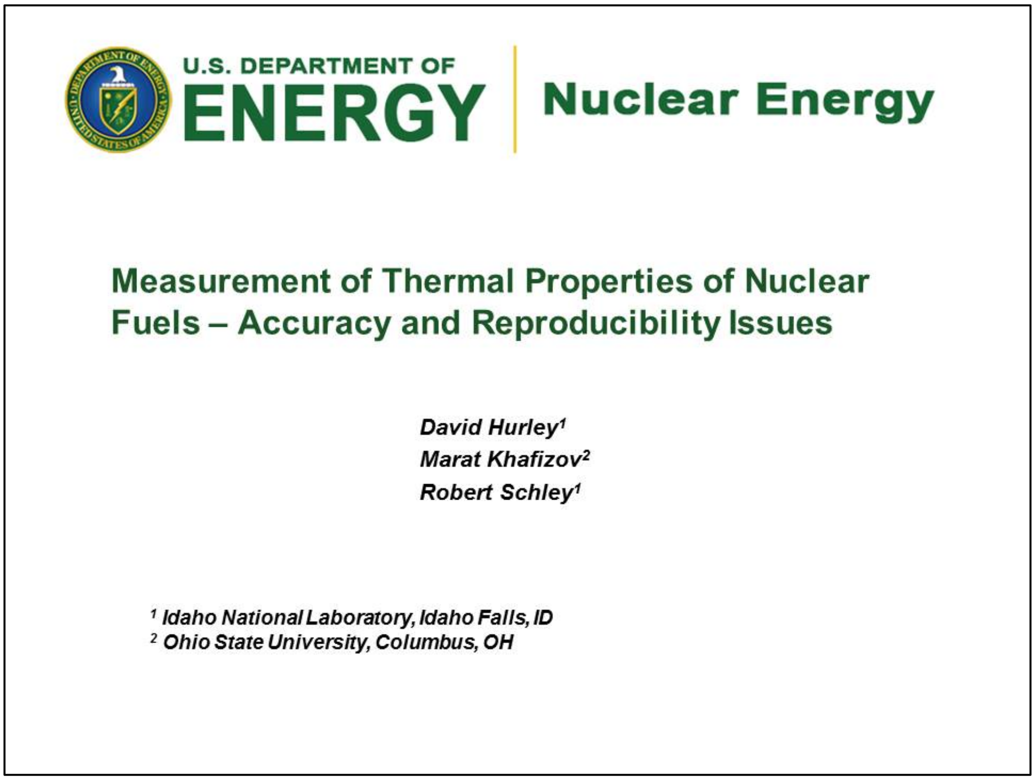
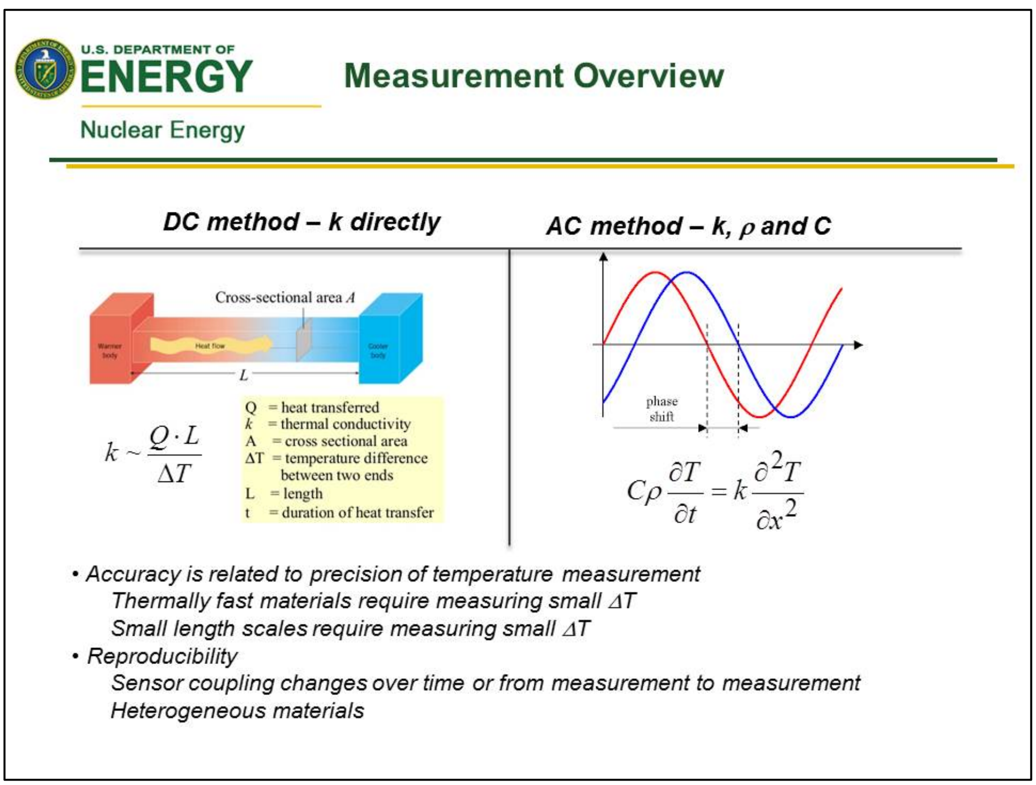
Chris	Stanek	505-664-0361	stanek@lanl.gov	CASL	LANL
David	Swank	208-526-1698	w.swank@inl.gov	Materials Science & Eng	INL
Mike	Tonks	208 526-6319	michael.tonks@inl.gov	Fuel Modeling & Simulation	INL
Dan	Wachs	208-526-7604	daniel.wachs@inl.gov	AFC Transient Testing R&D Lead	INL
Richard	Williamson	208-526-0576	richard.williamson@inl.gov	Fuels Modeling & Simulation	INL

Appendix C: Measurement of Thermal Properties of Nuclear Fuels – Accuracy and Reproducibility Issues – D. Hurley





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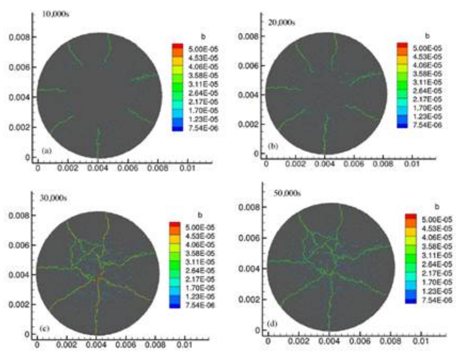
Issues Specific to Ceramic Nuclear Fuel

Simulated Cracks - Ceramic Fuel



H. Huang et al. / Nuclear Engineering and Design 278 (2014) 515–528

- *Heterogeneous – Ideally measurement length scale >> or << than heterogeneity length scale*
- *Heterogeneity length scale in ceramic fuel due to restructuring and cracking span from microns to millimeters*
- *Due to small diameter pellets and extensive cracking, sample can only be prepared with sizes /shapes which vary*
- *Must control atmosphere for elevated temperatures*
- *Sensitive components must be shielded - sample loading will not be trivial*
- *Measurements should be made in radial direction*

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Poor Candidates for Nuclear Fuel

- **Methods based on Franz Wiedemann Law – $k/\sigma=kT$**
  - L varies from metal to metal and is slightly temperature dependent for intermediate temps
  - Only measures electronic contribution
  - Phononic contribution for alloys can make a substantial contribution to overall conductivity
  - Doesn't work for phonon conductors
- **Scanning Thermal Microscopy (SThM)**
  - Uses AFM cantilevers in the form of a thermocouple or bolometer to map temperature field
  - Due to multiple modes of thermal coupling this method is only well suited to measure relative changes in conductivity
- **Raman Thermography**
  - Variation in Raman spectrum with temperature is used as a thermometer
  - Only works for Raman active materials like UO_2

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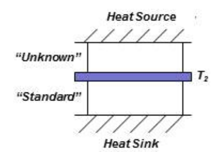
Potential Candidate – DC methods

- **DC methods measure conductivity directly**
- **Method is simple to apply (both data analysis and instrumentation)**
- **Typically requires large samples**
- **Variable coupling can cause serious reproducibility issues – especially for small samples**
- **Commercial systems exist**
- **PPMS from Quantum Design – 8mm thick puck for 2-50 W/mK range**
- **Modified plane wave method from Ctherm – diameter 17 mm**
- **Can develop old comparative measurement method for small samples**
  - RSI 21, 905, "A Method for Measuring the Thermal Conductivity of Small Samples of Poorly Conducting Materials Such as Optical Crystals"

C-Therm TCi
(Modified Transient Plane Source)

Thermal Transport Sample Puck PPMS



RSI, 21, 905, 1950

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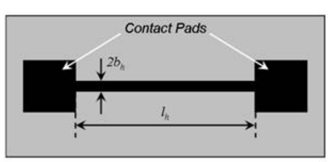
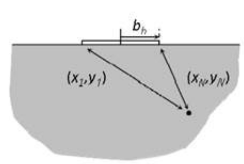
Potential Method - 3ω

- **Thin metal strip evaporated on the sample acts as heat source and a thermometer**
- **Voltage drop across heater at 3ω due to nonlinearity in resistive element's transfer function – R~(1+βΔT)**
- **Measures conductivity directly**
- **Strict geometry considerations must be realized – metal strip height, width, sample thickness and thermal penetration depth**
- **Requires separate measurement of β - larges source of error**
- **Location of measurement must be pre-selected before metal strip fabrication**





From Thesis by David Koninck, McGill University - Left: Schematic of the metal line filament deposed on a specimen for 3 ω measurement, Right: modling element with finite width.

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Potential Candidate – Laser Flash

- **Laser flash uses a long pulse laser or flash lamp (τ~ ms) and IR detector on back side of sample**
- **Requires separate measurement of specific heat to obtain condutivity**
- **Industry standard for measuring conductivity**
- **Development that spans 50 years of research**
- **Commercial systems have stringent requirements for sample size – to ensure measurement accuracy**
- **Size > 6 mm diameter and ~ 500 μm thickness (depends on conductivity range)**
- **Biggest issue with application to nuclear fuels is that the diameter requirement will be difficult if not impossible to meet for cracked fuel**
- **Cracking will not only result in irregular sizes and shapes but will result in laser flash-by**
- **ITU has designed custom laser flash system for nuclear fuel samples**
  - Laser flash-by and response of holder limit sample size
  - Accommodating smaller samples requires optical focusing of heating laser
  - More elaborate data analysis and more uncertainty

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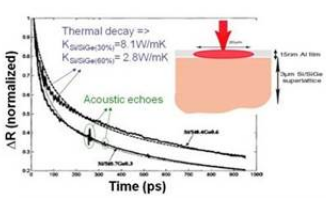
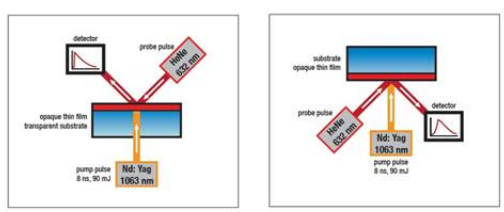
Potential Candidate – Time Domain Thermoreflectance

- **Heat pulse technique similar to laser flash but with much shorter heat pulses**
- **Micron lateral and 100nm – 1 μm depth resolution**
- **Requires coating sample with thin transducer film**
- **For un-calibrated films, measures thermal effusivity and Kapitza resistance**
- **Research systems typically involve ultrafast lasers**
- **Commercial system available from Linseis that uses ns laser**
- **Primary advantage is that it can access small length scales**



Y. Ezzahri et al. Appl. Phys. Let. 2005



TDTR system from Linseis

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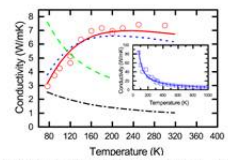
Potential Candidate – Frequency Domain Methods

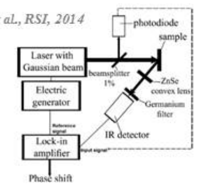
- **Change sampling volume by changing source size and modulation frequency**
- **No commercial system exist**
- **Can be divided into two sub-categories – both use laser heating**
  - Thermoreflectance – monitors small temperature induced changes in reflectivity
  - Photothermal radiometry – monitors blackbody radiation from either front or back surface
- **Thermoreflectance**
  - Single side access
  - Sampling volume varied from 10 μm^3 to > 100 μm^3
  - Typically require coating sample with thin film
  - Can measure conductivity and diffusivity directly
  - Thermal Conductivity Microscope



Journal of the American Ceramic Society—Khafizov et al.

- **Photothermal radiometry**
  - Single side access
  - Sampling volume > 100 μm^3
  - Not required to coat sample with film
  - Can measure diffusivity

Pham et al., RSI, 2014



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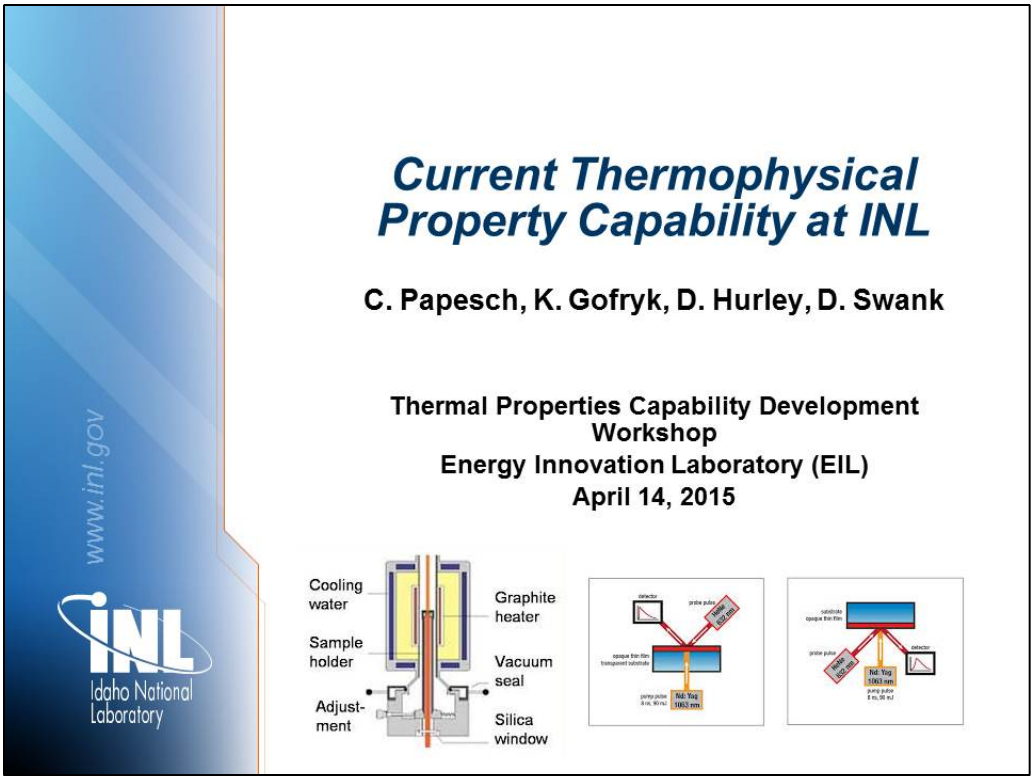
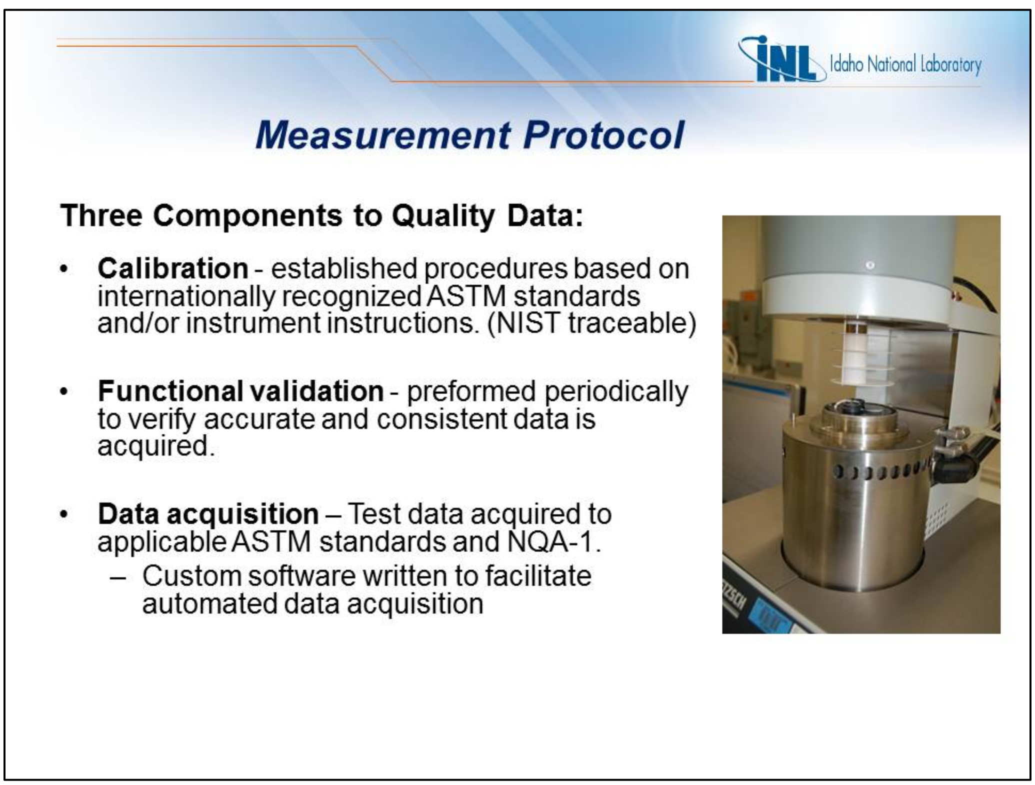
Conclusions

- **One size solution for measurement of conductivity does not exist**
- **There are a number of experimental methods to measure thermal conductivity**
- **Each method is suitable for a limited range of materials and geometries**
- **Measurements of nuclear fuel introduce significant challenges**

Appendix D: Current Thermophysical Property Capability at INL – C. Papesch





Material Characterization Area
East room of FASB - MFC

- Set up to measure HEU materials and irradiated non-fuel materials with low activity
- Measurements available
  - Bulk density
  - Thermal Expansion, CTE
  - Specific Heat
  - Thermal diffusivity
- Sample preparation is conducted in fume hoods / gloveboxes


Fresh Fuels Glovebox Laboratory
room B127 Analytical Laboratory - MFC

- Set up to measure Transuranic materials and irradiated non-fuel materials with low activity
- Measurements available
  - Optical microscopy
  - Thermal Expansion, CTE
  - Specific Heat
  - Thermal diffusivity
- Other capabilities
  - Analytical balance
  - Annealing furnace

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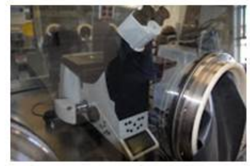
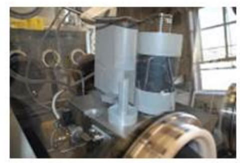
Fresh Fuels Glovebox Laboratory

room B127 Analytical Laboratory - MFC

Measurement	Instrumentation
Mass	Mettler Toledo
Thermal Diffusivity	Netzsch LFA 427
CTE	Netzsch DIL 402 C
Specific Heat	Netzsch DSC 404
STA/TGA	Netzsch STA F1 Jupiter
Optical Microscope	Leica DM5000i





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INL Carbon Characterization Laboratory (CCL)

(located in Lab C-19 and C-20 of the IRC)

Currently, instrumentation, fixtures and methods are in place for pre and post irradiation material property measurements of:

- Bulk density
- Thermal diffusivity
- CTE
- Elastic modulus
- Electrical resistivity
- Specific Heat.

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Rad Material Capable Equipment

Note: Rad capable refers to the ability to handle low activity specimens.
(~<50 mRem/hr β-γ on contact) (examples: DU or slightly activated materials)

Measurement	Instrumentation
Mass	2 ea. Sartorius Balance ME235P 2ea.
Thermal Diffusivity	Netzsch LFA 457 2ea
CTE	Netzsch DIL 402 C 2ea.
Electrical Resistivity	Four point probe technique
Elastic Modulus	Nondestructive by sonic velocity and/or sonic resonance
Specific Heat	Netzsch DSC 404C
STA/TGA	Netzsch STA 449C

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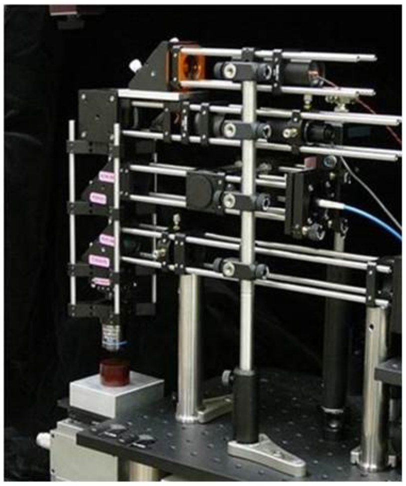
Rad Material Capable Equipment (cont.)

- Two position glove box (HEPA filtered)
- Fume Hood (HEPA filtered)
- LASER engraver (HEPA filtered)
- Tensile/compression test frame
- Optical microscope
- Photography
- ThermoTech High Temperature Furnace (2500°C)

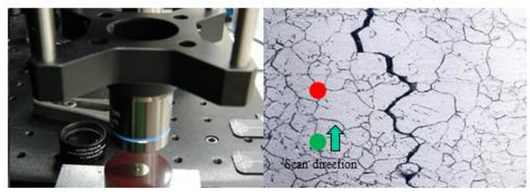
Thermal Conductivity Microscope (TCM)

Prototype System



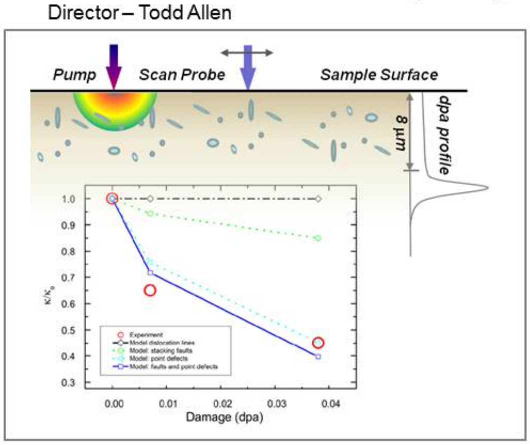
- The TCM uses lasers to locally heat and map temperature field
- Currently a prototype TCM has been constructed and is undergoing testing
- Used to measure UZr, UO2 and UN samples
- 10 – 100 μm field of view complements LFA

Tight Optical Focusing and Imaging



History of TCM Development

Center for Materials Science of Nuclear Fuel (FY10-15)
Director – Todd Allen



Journal of Nuclear Materials (2014), doi: http://dx.doi.org/10.1016/j.jnucmat.2014.07.053

- The development of the TCM started under the Center for Materials Science of Nuclear Fuel – an Energy Frontier Research Center based at INL
- Originally the TCM was used to measure the reduction in thermal conductivity of UO2 that had been ion irradiated
- The spatial resolution of TCM is ideally suited to measure thin irradiation layers
- The TCM was also used to measure thermal transport across individual grain boundaries
- The emphasis of the CMSNF was to connect irradiation microstructure to thermal conductivity in UO_2

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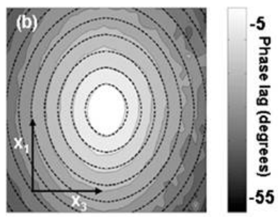
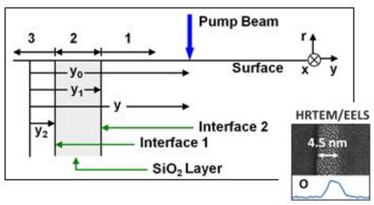
Specifications for the TCM

TW profile for quartz



15 µm × 15 µm

Transport Across Grain Boundary



- Small sample sizes (100 µm^3 and larger)
- Sampling volume varied from 10 µm^3 to 100 µm^3
- Can be used to measure diffusivity and conductivity
- Does not require calibration
- Only requires single-side access
- Can be used to image thermal anisotropy directly
- Sample preparation requirements on no more restrictive than laser flash
- Does not cover the 100 µm to 1 mm gap
- Typically requires sample to be coated with film
- Low temperature range – 10 K to room temperature
- High temperature range – preliminary results up to 800 C

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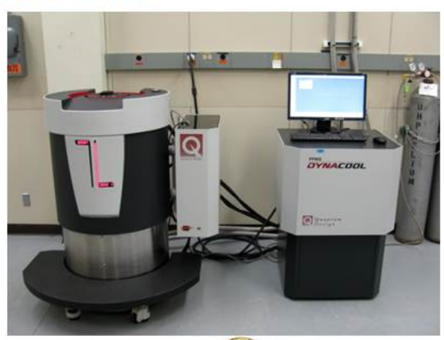
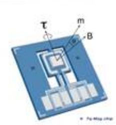
Physical Properties Measurement System (DynaCool-9 PPMS)

Laboratory (IF-603, C6)

- **State-of-the-are closed circle measurement system (He gas)**
- **Measurement platform:**
  - **Temperature range:**
    - **T = 1.8 – 400 K**
  - **Magnetic field range:**
    - **H = 0 – 9 T**
  - **Sample rotation:**
    - f = 0 – 360º



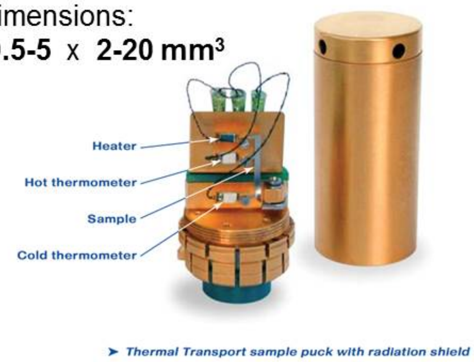


- **Measurements options:**
  - ac/dc resistivity, ac/dc magnetic susceptibility, magnetic torque, STM, Hall effect, **Seebeck effect**, **heat capacity, thermal conductivity**, … and many more !

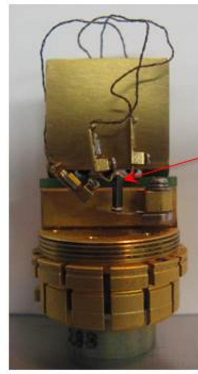
Thermal conductivity: PPMS

"one heater two thermometers method"

Sample dimensions:
0.5-5 x 0.5-5 x 2-20 mm^3



Typical assembly:



UO_2 <110>

- **Pulse-power (Maldonado) method:**

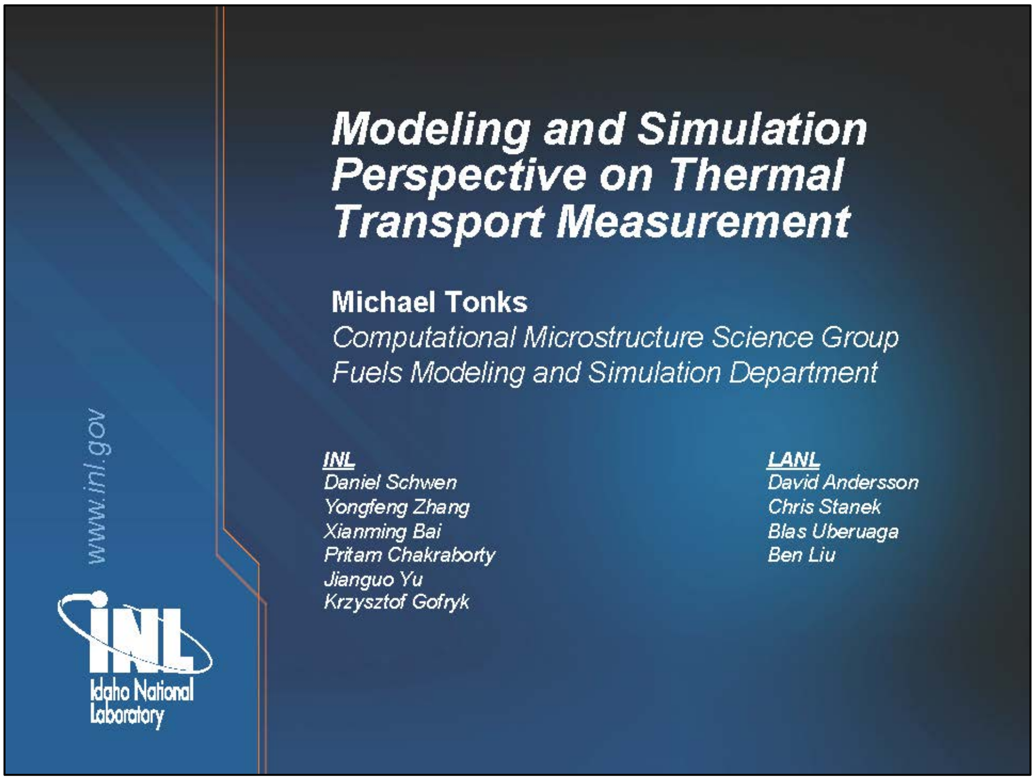
$$\Delta T_{model} = \Delta T_{\infty} \times \{1 - [\tau_1 \times \exp(-t/\tau_1) - \tau_2 \times \exp(-t/\tau_2)]/(\tau_1 - \tau_2)\}$$

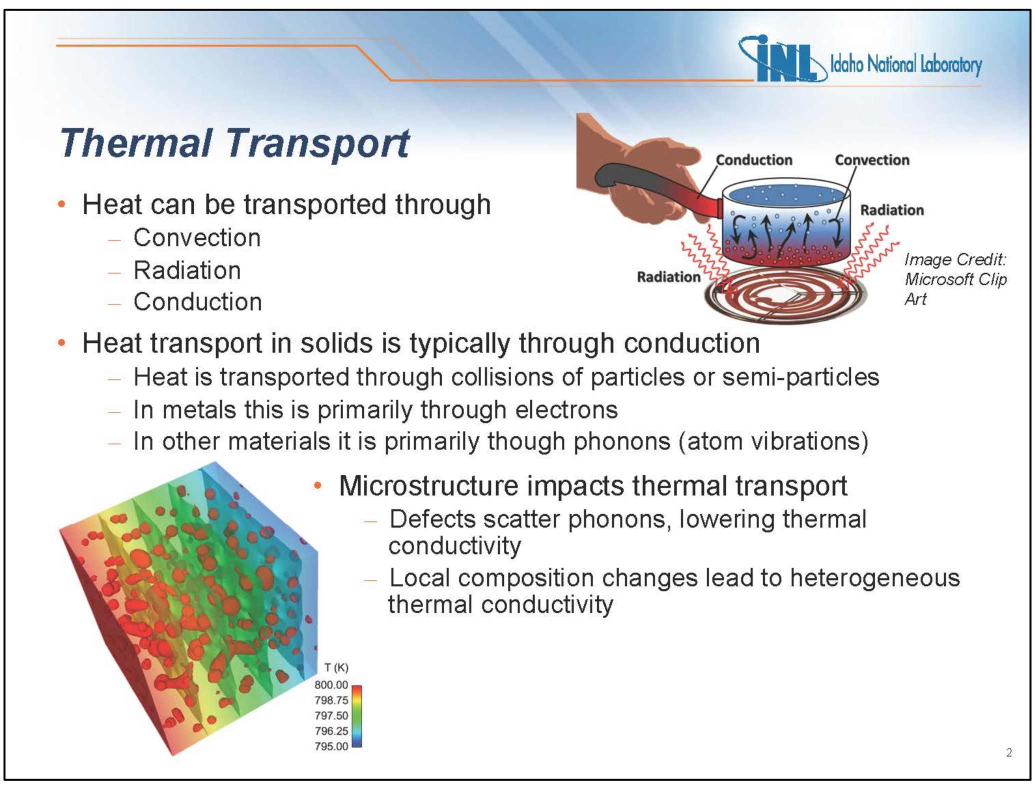
- **Direct (steady-state) method**: $K = P/\Delta T$

Facility Summary

Laboratory / Instrument	Facility	Materials	Approximate Radiological values
Materials Characterization	FASB – MFC	HEU / irradiated non-fuel	100 mR/h βγ
Fresh Fuels Glovebox	AL-MFC	TRU / irradiated non-fuel	500 mR/h βγ
ICCL	IRC	DU / irradiated non-fuel	<50 mR/h βγ
TCM	IRC	DU / irradiated non-fuel	<50 mR/h βγ
PPMS	IRC	Non-radiological	n/a

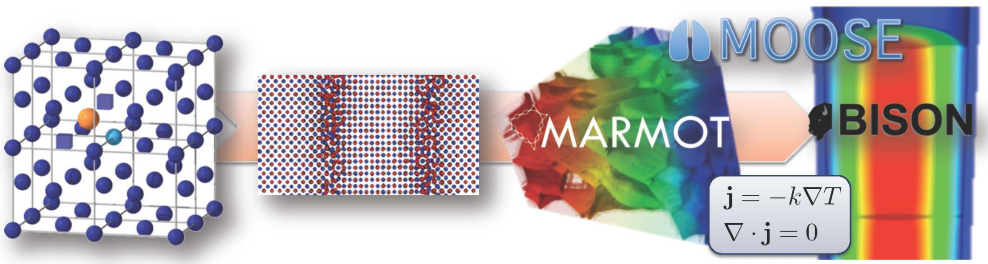
Appendix E: Modeling and Simulation Perspective on Thermal Transport Measurements – M. Tonks





Multiscale Simulation of Heat Transport

- Heat transport can be simulated with various approaches
- Smaller scale simulations can capture more physical phenomena but are limited on size of domain



nanometers
First Principles
- Use Boltzmann transport equation
- Inputs provided by simulations

100's of nanometers
Molecular Dynamics
- Explicitly model phonon transport
- Can't model electron transport

microns
Mesoscale
- Solves Fourier's law with microstructure dependent thermal conductivity

millimeters and up
Engineering scale
- Solves Fourier's Law

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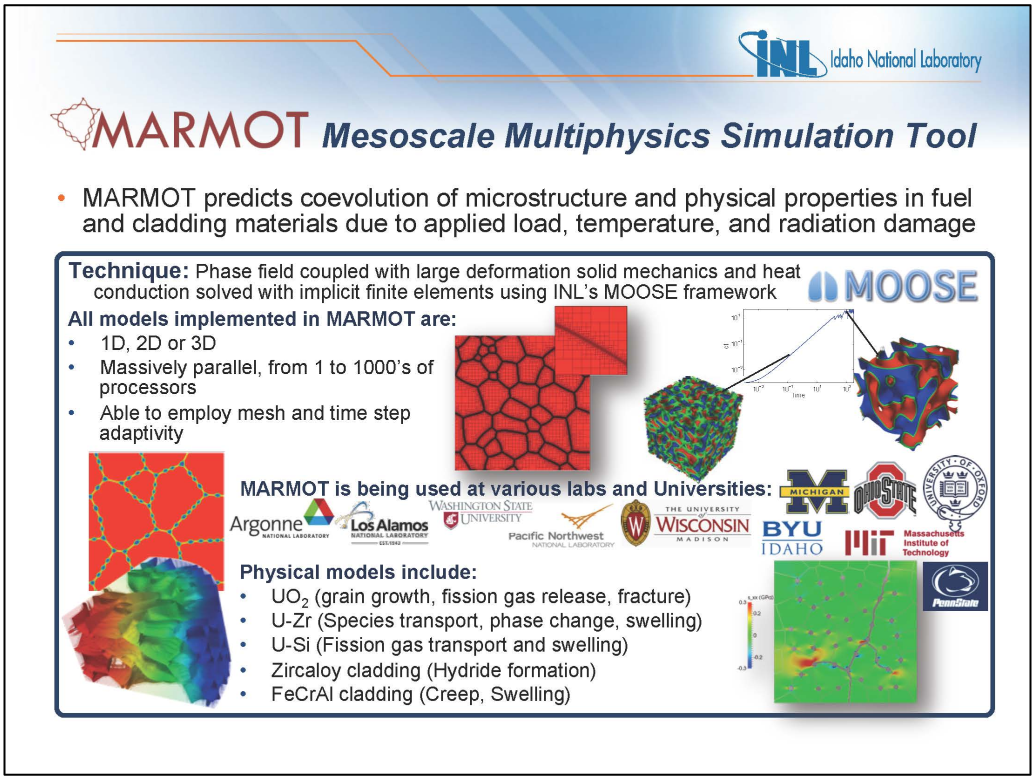
Thermal Conductivity

- The thermal conductivity plays a critical role in Fourier's law, as it defines how efficiently heat can be conducted through a material
- The thermal conductivity must account for the impact of microstructure and defects below the resolution of your problem
- Thermal conductivities can be measured or calculated with lower length-scale simulation

DFT and Boltzmann Transport Equation
- Can account for phonon transport or electron transport
- Can account for various scattering mechanisms of perfect crystals or small defects
- Requires 100's of DFT calculations for higher order descriptions

Molecular Dynamics
- Can only account for phonon transport
- Can account for scattering mechanisms due to various defects, including point defects, dislocations, and GBs
- Depends on empirical potential accuracy
- Limited length-scale

Mesoscale Solution of Fourier Equation
- Only accounts for changes in local thermal conductivity
- Requires that the local thermal conductivity be known in each microstructural feature
- Can model 100's of microns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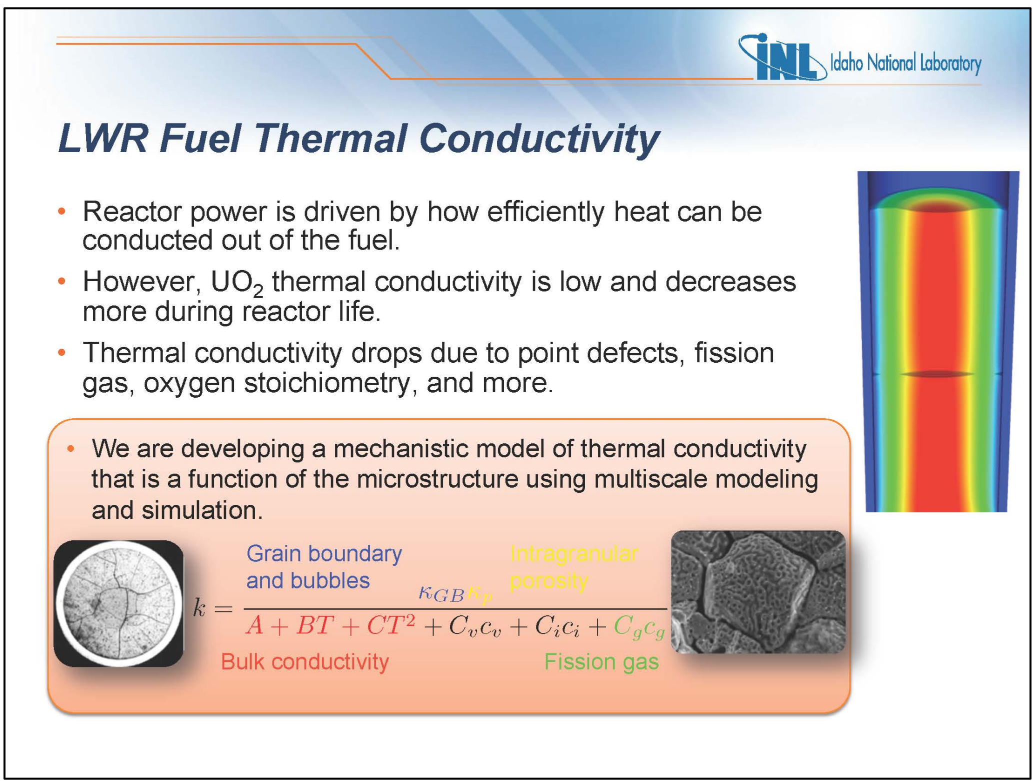
MARMOT Mesoscale Multiphysics Simulation Tool
• MARMOT predicts coevolution of microstructure and physical properties in fuel and cladding materials due to applied load, temperature, and radiation damage
Technique: Phase field coupled with large deformation solid mechanics and heat conduction solved with implicit finite elements using INL's MOOSE framework
MOOSE
All models implemented in MARMOT are:
• 1D, 2D or 3D
• Massively parallel, from 1 to 1000's of processors
• Able to employ mesh and time step adaptivity
MARMOT is being used at various labs and Universities:
Argonne National Laboratory
Los Alamos National Laboratory
Washington State University
Pacific Northwest National Laboratory
The University of Wisconsin Madison
BYU Idaho
Michigan
Ohio State
Massachusetts Institute of Technology
PennState
Physical models include:
• UO_2 (grain growth, fission gas release, fracture)
• U-Zr (Species transport, phase change, swelling)
• U-Si (Fission gas transport and swelling)
• Zircaloy cladding (Hydride formation)
• FeCrAl cladding (Creep, Swelling)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LWR Fuel Thermal Conductivity
• Reactor power is driven by how efficiently heat can be conducted out of the fuel.
• However, UO_2 thermal conductivity is low and decreases more during reactor life.
• Thermal conductivity drops due to point defects, fission gas, oxygen stoichiometry, and more.
• We are developing a mechanistic model of thermal conductivity that is a function of the microstructure using multiscale modeling and simulation.
Grain boundary and bubbles
Intragranular porosity
$$k = \frac{\kappa_{GB}\kappa_p}{A + BT + CT^2 + C_v c_v + C_i c_i + C_g c_g}$$
Bulk conductivity
Fission gas

$k = \frac{\kappa_{GB}}{A + BT + CT^2 + C_v c_v + C_i c_i + C_g c_g}$

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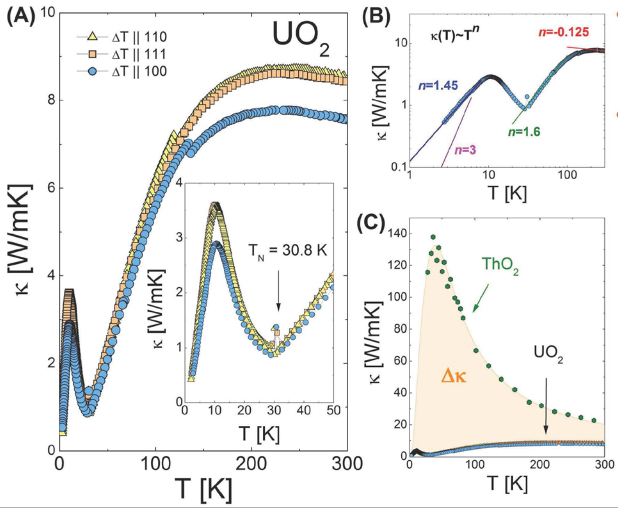
Single Crystal UO_2 Thermal Conductivity

Los Alamos NATIONAL LABORATORY EST. 1943

- The UO_2 thermal cond. is unusually low, and is over-predicted by MD
- This discrepancy appears to be due to magnetic spin scattering



- Spin scattering also leads to anisotropy in the thermal conductivity
- Comparison to ThO_2 (no f electrons or magnetism) is very enlightening:
  - Max(κ_{ThO2}) is 140 W/mK at 40 K.
  - Max(κ_{UO2}) is 8.7 W/mK at 220 K.

K. Gofryk et al. Anisotropic thermal conductivity in uranium dioxide, Nature Comm. 2014, DOI: 10.1038/ncomms5551

7

$k = \frac{\kappa_{GB}}{A + BT + CT^2 + C_v c_v + C_i c_i + C_g c_g}$

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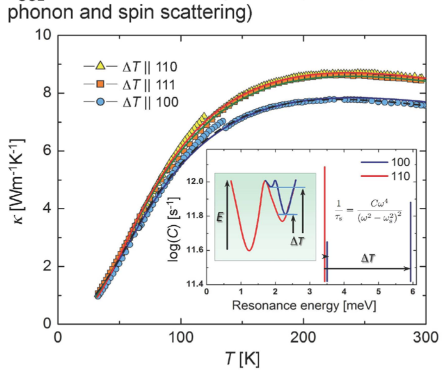
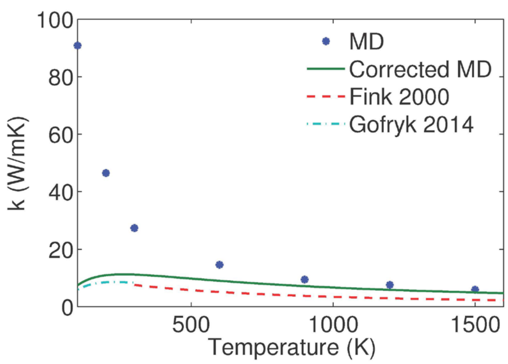
Correcting κ_{UO2} predicted by the MD simulations

- MD cannot capture the spin scattering mechanism directly.
- Therefore, we derive the spin scattering contribution from experiments and correct the simulations.

κ_{UO2} fitted to the Callaway model (defect, boundary, phonon and spin scattering)



MD simulations corrected for spin scattering



Los Alamos NATIONAL LABORATORY EST. 1943

$$k = \frac{\kappa_{GB}}{A + BT + CT^2 + C_v c_v + C_i c_i + C_g c_g}$$

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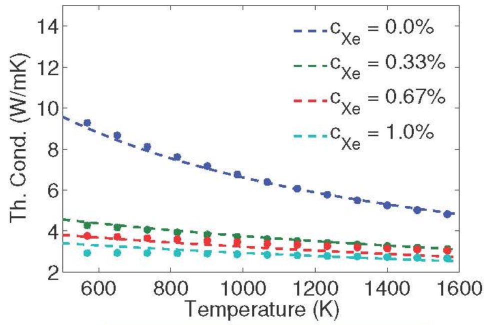
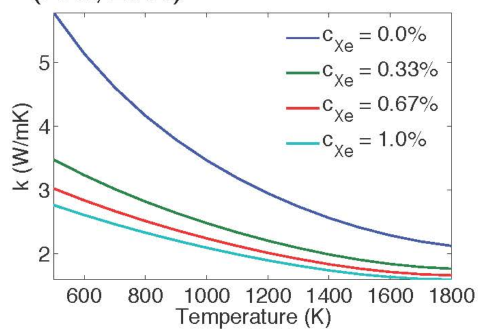
Development of Correction for Dispersed Xe

- Additional phonon scattering due to Xe is quantified using molecular dynamics to determine correction
- The MD simulations are corrected to account for spin scattering
- The corrections were then added to the thermal conductivity equation fit from experiments (Fink, 2000)





Fit correction: $\ldots + 1.5c_g^{0.45}$

Los Alamos NATIONAL LABORATORY EST. 1943

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$$k = \frac{\kappa_{GB}\kappa_p}{A + BT + CT^2 + C_v c_v + C_i c_i + C_g c_g}$$

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Intragranular Bubble Contribution

UF UNIVERSITY of FLORIDA

- Intragranular bubbles in the fuel are small and spherical, $r_b < 5$ nm.
- Various analytical models exist that represent porosity using a linear thermal resistor approximation
- MD simulations have shown that phonon scattering must be accounted for to accurately represent small bubbles

Maxwell-Eucken (no phonon scattering)

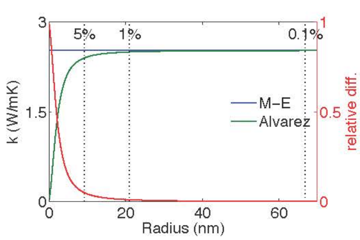
$$\kappa_{ME} = \frac{1-p}{1+p/2}$$

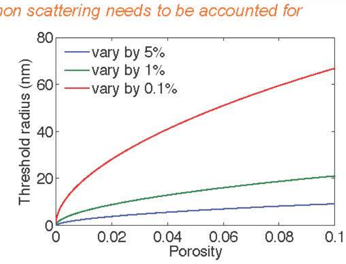
vs

Alvarez (phonon scattering)

$$\kappa_A = \left(\frac{1}{k_{ME}} + \frac{9l^2}{2r^2}\frac{p(1+3\sqrt{p/2})}{1+(0.864+0.29e^{-1.25\frac{r}{l}})\frac{l}{r}}\right)^{-1}$$

Phonon scattering needs to be accounted for





C.-W. Lee, et al., J. Nucl. Mater., 456 (2015) 253 – 259

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$k = \frac{\kappa_{GB}}{A + BT + CT^2 + C_v c_v + C_i c_i + \dots}$

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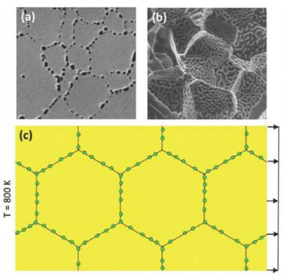
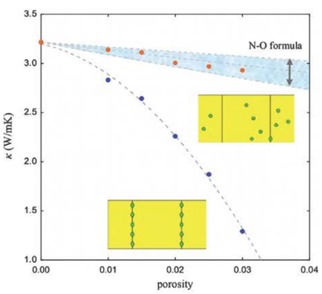
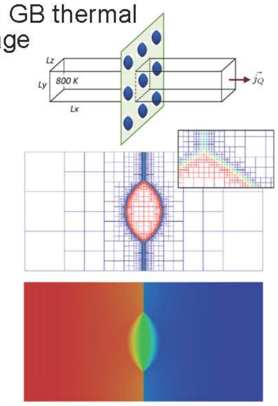
Grain Boundary Bubble Contribution

- Mesoscale solutions of the Fourier equation have shown that bubbles aligned on GBs have a larger impact on the thermal conductivity than randomly distributed bubbles
  - They can be accounted for by changing the effective GB thermal resistance R' to be a function of the fractional coverage







- Thus, a mechanistic model is needed that accounts for this effect.

Millett and Tonks, J. Nucl. Mat., 412:3 (2011) 281–86 Millett, et al., J. Nucl. Mater. 439 (2013) 117–122. 11

$k = \frac{\kappa_{GB}}{A + BT + CT^2 + C_v c_v + C_i c_i + \dots}$

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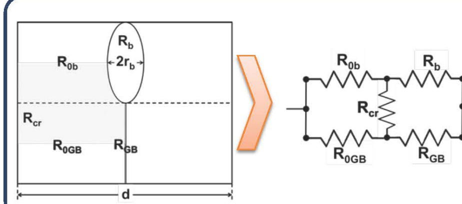
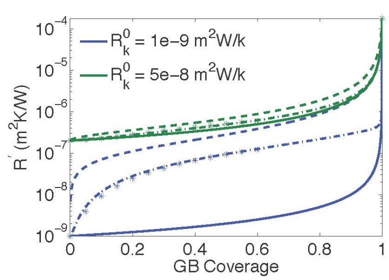
Model of Grain Boundary Bubble Contribution

- A thermal resistor model is created to describe the impact of GB bubbles on the thermal conductivity



- System is represented by one bubble and one section of GB
- It is approximated by five thermal resistors
  - Three for bulk UO_2
  - One for bubble
  - One for GB

- $R_{cr} = 0$ gives a lower bound of the GB thermal resistance
- $R_{cr} = large$ gives an upper bound
- By fitting to 3D MARMOT simulation results we obtain:
  - $R_{cr} = 5\times10^{-7}$ m²W/K



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$$k = \frac{\kappa_{GB}\kappa_p}{A + BT + CT^2 + C_v c_v + C_i c_i + C_g c_g}$$

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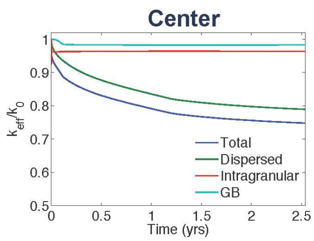
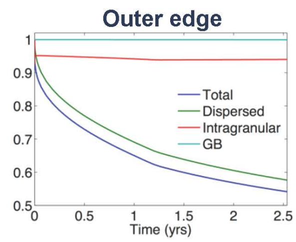
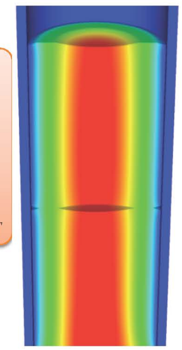
Investigation of Fission Gas Th. Cond. Model

- To investigate the relative importance of the three distributions of fission gas, we outputted the fission gas variables from a BISON simulation from a ten pellet rodlet at two locations:
  - Pellet center
  - Outer edge





Outputs:
- Grain size
- Dispersed fission gas c_g
- Intragranular porosity p_{ig}
- Fractional coverage f_c
- Temperature T



- Effect of dispersed fission gas is much higher than the intragranular bubbles and the GB bubbles

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$$k = \frac{\kappa_{GB}\kappa_p}{A + BT + CT^2 + C_v c_v + C_i c_i + C_g c_g}$$

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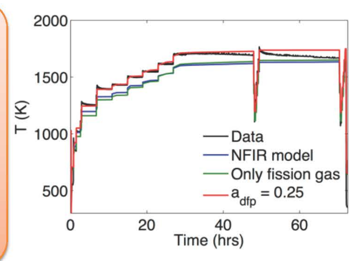
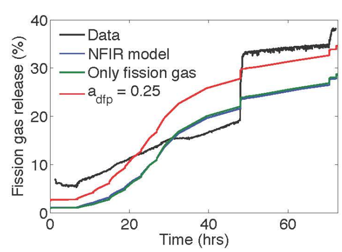
Implementation in BISON

- Our mechanistic model accounts for the impact of fission gas on the thermal conductivity and it couples to the fission gas release model
- Until we have developed models for the other defects, we use a semi-empirical model from Lucuta, et al., 1996.
  - Irradiation damage contribution (point defects)
  - Precipitated fission products contribution
  - Dissolved fission products contribution, however this contribution also accounts for dissolved fission products.

- Fission gas is about 25% of dissolved fission products
- Its impact on the thermal conductivity is higher
- We remove the fission gas by only using a percentage of the factor from Lucuta, fit to RISO-3 AN3 assessment data





14

$$k = \frac{\kappa_{GB}\kappa_p}{A + BT + CT^2 + C_v c_v + C_i c_i + C_g c_g}$$

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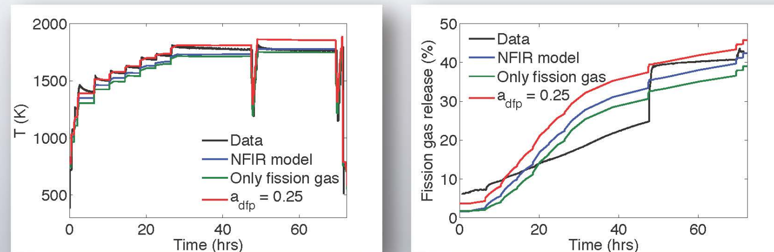
Comparison to Assessment Data

- To validate our mechanistic model, we compare the BISON predictions to assessment data.
- One such comparison is complete so far, but more are coming.

RISO-3 AN4 Assessment Case



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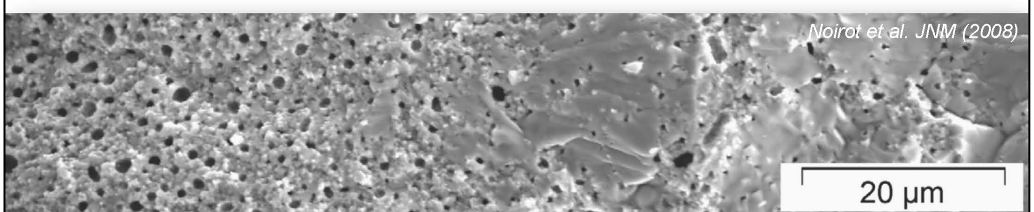
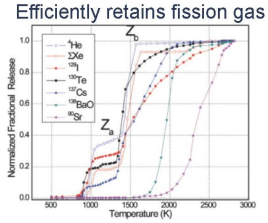
High Burnup Structure (HBS)



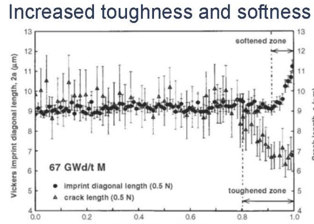
- HBS forms in fuel with burnup > 60 GWd/tHM and at temperatures < 1300 K. It has small grains (200 nm) and large bubbles (1 µm).
- Recent research shows that HBS has favorable properties.

Efficiently retains fission gas



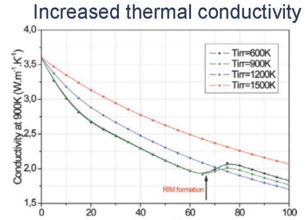
From Hiernaut et al. JNM (2008)

Increased toughness and softness



From Spino et al. JNM (2003)

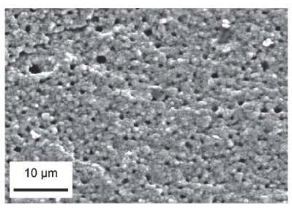
Increased thermal conductivity



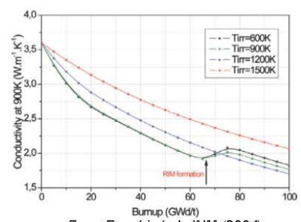
From Ronchi et al. JNM (2004)

Modeling Approach

- We are using the approach we have taken to develop a thermal conductivity model of fission gas to investigate why HBS has a higher thermal conductivity.
- There are two main goals
  - Determine if the microstructure alone is sufficient to account for the thermal conductivity increase
  - Determine the impact of dispersed fission gas on the HBS thermal conductivity



Rondinella et al. Materials Today (2010)



From Ronchi et al. JNM (2004)

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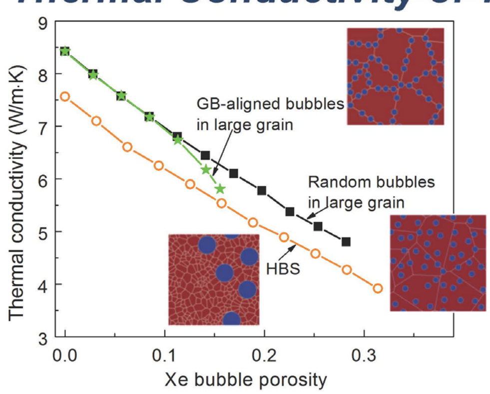
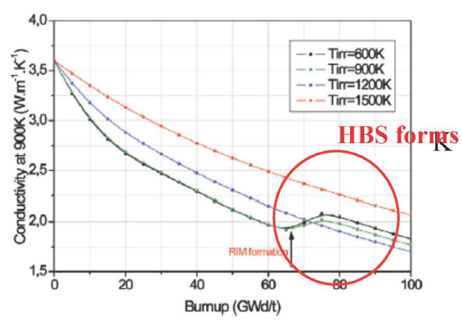
Thermal Conductivity of Three Microstructures





- Our simulations results find:
  - Aligned bubbles have more reduction than random bubbles at p > 0.1
  - HBS has a lower thermal conductivity due to high-density of GBs
  - Topology of microstructures cannot explain the increased thermal conductivity of HBS – some important physics is missing

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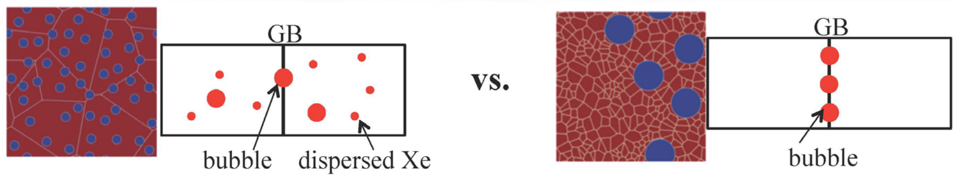
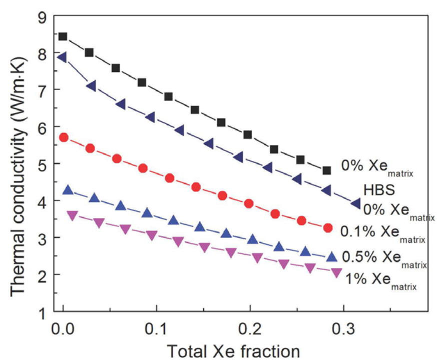
Effect of Dispersed Xe on Thermal Conductivity



Large grains, dispersed Xe, random bubbles **Small grains, big bubbles, no dispersed Xe**



- In HBS, all dispersed Xe is absorbed by GBs due to the high GB density
- In large-grain sample a small amount of dispersed Xe stays in the grain interior
- Only 0.1% dispersed Xe in the large-grain sample is sufficient to lower its thermal conductivity below HBS
- More dispersed Xe, more reduction

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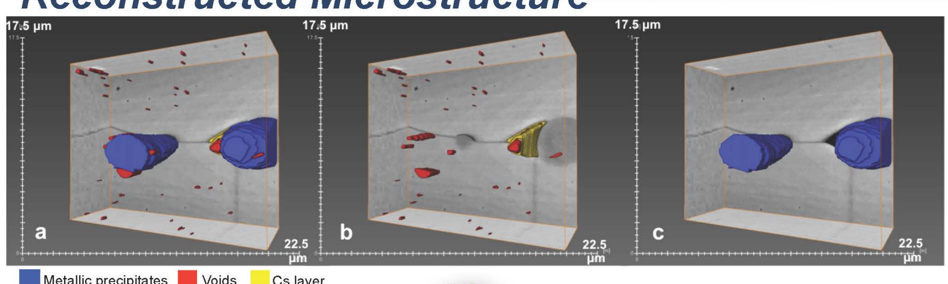
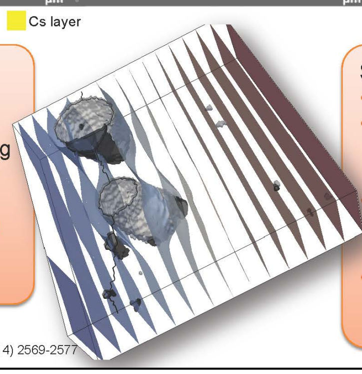
Thermal Conductivity Calculation On Reconstructed Microstructure



Experimental Setup
- Highly irradiated mixed oxide fuel was characterized in 3D using EBSD, EDS and FIB sectioning.
- More efficient sectioning approaches are under investigation

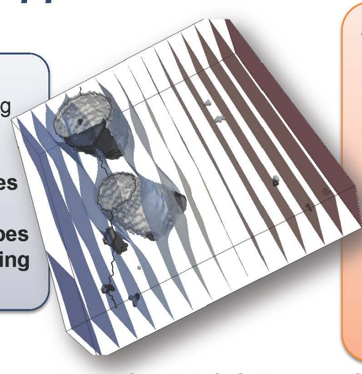


Simulation
- Microstructure was reconstructed
- The effective thermal conductivity was predicted using a steady state heat conduction simulation
- The thermal conductivity went up due to metallic precipitates when it should have gone down
- **Error was due to concentration of defects below the experimental resolution.**

Teague, Fromm, Tonks, Field. JOM, 66:12 (2014) 2569-2577

Alternative Approach #1

Experimental Work
- 3D characterization using EBSD, EDS and FIB
- **Use FIB to lift out TEM samples from all phases in the microstructure**
- **Characterize defect types and concentrations using TEM**



Simulation
- **Use DFT and MD simulations to quantify the impact of each defect type on the microstructure**
- **Develop equations that account for phonon scattering by each defect as a function of concentration**
- Use MARMOT to compute effective thermal conductivity
  - Microstructure from EBSD
  - Defect concentrations in each phase from TEM

- This approach uses experimental data on microstructure (EBSD) and defects (TEM) to inform the mesoscale calculation
- The final result would be more accurate, but would have significant uncertainty since the defect contributions would not be validated

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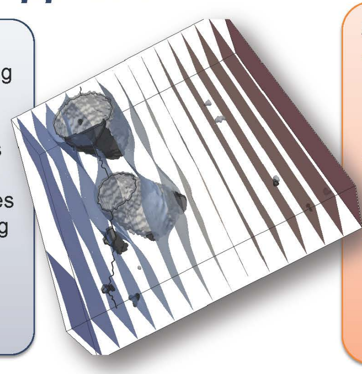
Alternative Approach #2

Experimental Work
- 3D characterization using EBSD, EDS and FIB
- Use FIB to lift out TEM samples from all phases in the microstructure
- Characterize defect types and concentrations using TEM
- **Measure local thermal conductivities of each phase**



Simulation
- Use DFT and MD simulations to quantify the impact of each defect type on the microstructure and **validate by comparing to data**
- Develop equations that account for defect phonon scattering as a function of concentration
- Use MARMOT to compute effective thermal conductivity
  - Microstructure from EBSD
  - Defect concentrations in each phase from TEM

- This approach uses experimental data on microstructure (EBSD) and defects (TEM), as well as thermal conductivity to inform the mesoscale calculation
- The final prediction would be accurate and validated, due to detailed microstructure characterization and thermal conductivity measurement at the microstructure level

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Experimental Needs

- We need as much data as we can get
- Our wish list would be
  - Detailed characterization of the microstructure at every scale that impacts the thermal conductivity
    - Defect types, concentrations, and size distributions
    - Microstructure characterization including structure, orientation, and composition
  - Information about the thermal conductivity of each phase
    - Single crystal measurements
    - Measurements of GB thermal resistance
    - Estimates of the impact of defect on the phase conductivities
- However, bulk thermal conductivity measurements are still very useful for validation at the macroscale (BISON)

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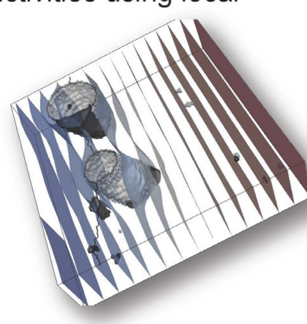
Validation Needs

- The thermal conductivities calculated with atomistic methods need to be validated
  - Single crystal thermal conductivities
  - Single crystal conductivities with different defect concentrations
  - GB thermal resistance measurements
- The approach to calculate effective thermal conductivities using local conductivities needs to be validated
  - Local thermal conductivity measurements of a well characterized microstructure
  - Bulk thermal conductivity measurements of the same or similar sample
  - We will then directly compare the computed effective thermal conductivity (using the measured local conductivities) to the bulk measurement



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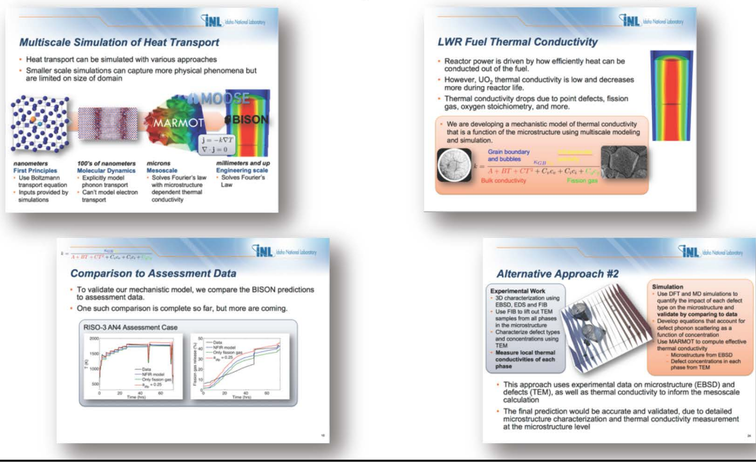
Conclusions

- Modeling and simulation are powerful tools for investigating thermal transport in nuclear materials
- Simulation approaches across length-scales have specific benefits and weaknesses, and thus are most powerful when used together



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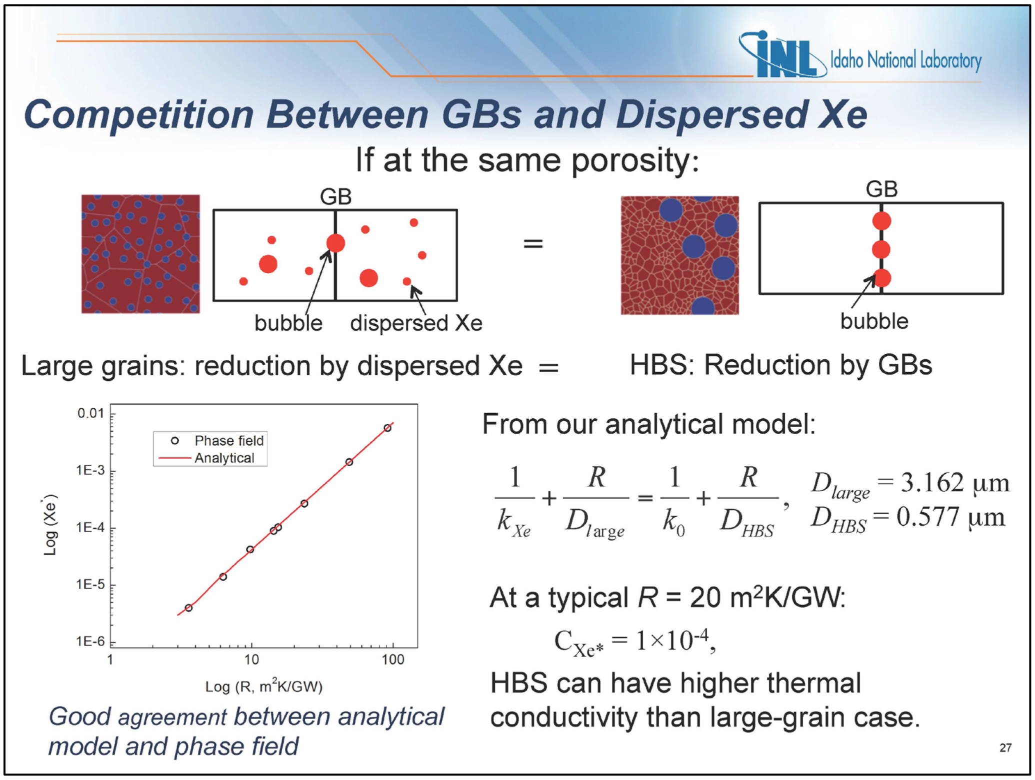
For more information, contact Michael Tonks at michael.tonks@inl.gov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Competition Between GBs and Dispersed Xe
If at the same porosity:
GB
bubble
dispersed Xe
=
GB
bubble
Large grains: reduction by dispersed Xe =
HBS: Reduction by GBs
Phase field
Analytical
0.01
1E-3
1E-4
1E-5
1E-6
1
10
100
Log (Xe*)
Log (R, m²K/GW)
Good agreement between analytical model and phase field
From our analytical model:
$\frac{1}{k_{Xe}} + \frac{R}{D_{large}} = \frac{1}{k_0} + \frac{R}{D_{HBS}}$, $D_{large} = 3.162\ \mu m$, $D_{HBS} = 0.577\ \mu m$
At a typical R = 20 m^2K/GW:
$C_{Xe*} = 1\times10^{-4}$,
HBS can have higher thermal conductivity than large-grain case.
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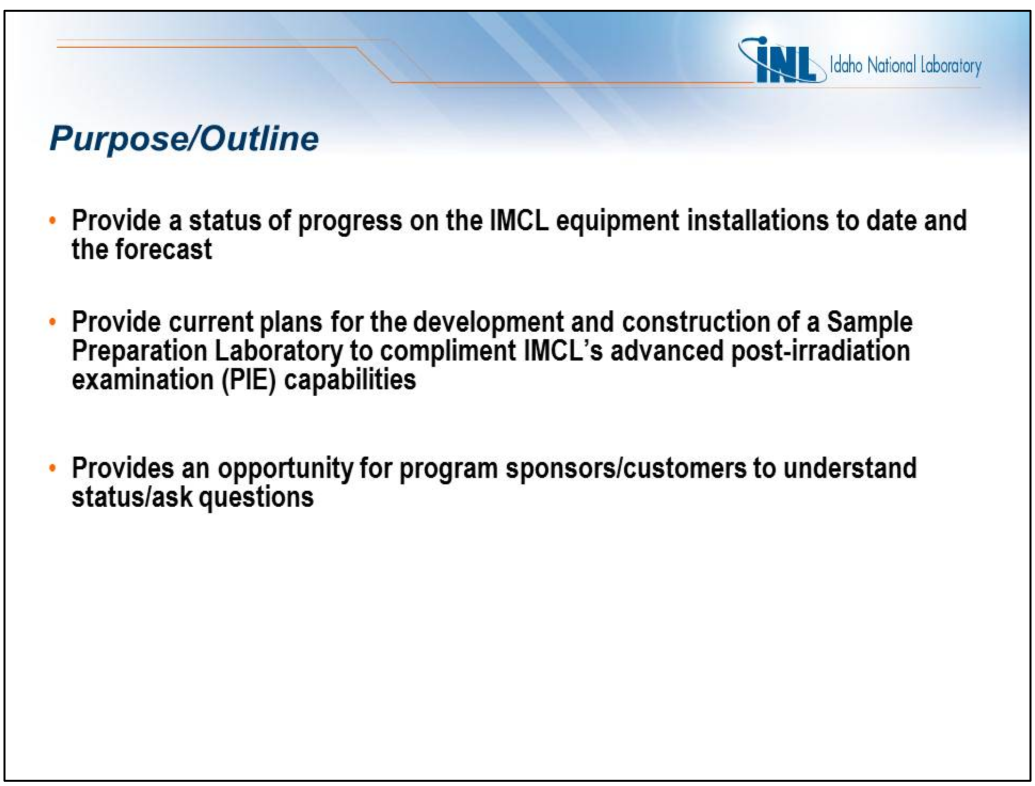
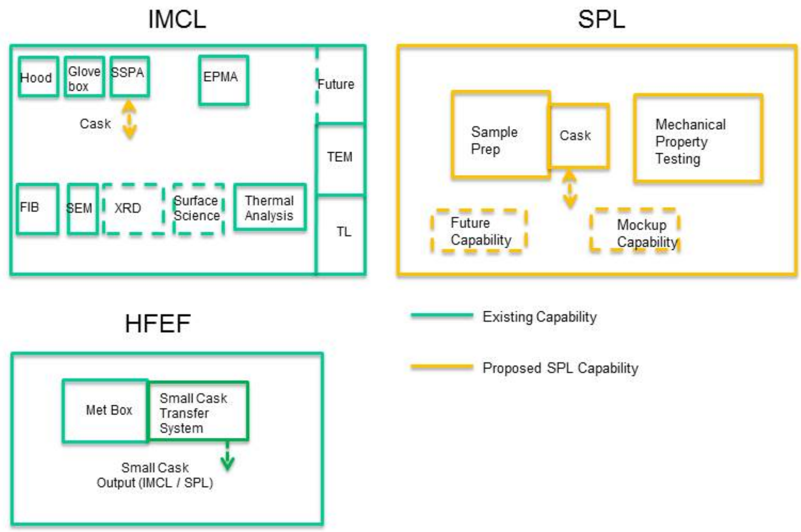
Appendix F: *Irradiated Materials Characterization Laboratory (IMCL) Equipment Installation and Sample Prep Laboratory (SPL) Status & Future Plans – C. Knight*



Advanced Post Irradiation Examination Capability Vision



IMCL Equipment Installation

- **The IMCL facility represents a new innovative approach to R&D on high dose rate materials.**
  - Carefully integrating advanced, high resolution microscopy equipment with shielding and confinement systems in a new and unique way - and integrates facility operations and equipment installations in parallel
- **Goal is to get the most use of each piece of equipment at the earliest opportunity, while maximizing facility functionality & maintaining safe and proficient conduct of operations.**
- **Through FY18 new equipment is being installed and made operational, with equipment capabilities transitioning from work on cold samples to high dose rate samples as experience is gained on equipment**
- **Original planning assumed yearly allotments to complete installations – however, $18M was provided in FY15 budget to fund remaining equipment installations**
  - Move up TEM Isolation Room from FY 2017 to FY 2016
  - Move up Thermal Properties Cell from FY 2018 to FY 2017
  - Move up Mechanical Properties Test Cell from FY 2018 to 2017

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Current IMCL Status

- Completed initial facility readiness review in FY 14 – facility "operational".
- Shielded Sample Preparation Area (SSPA) installed – (additional work in progress as outlined in next slide).
- Electron Probe Micro Analyzer (EPMA) relocated from Analytical Lab and installed
- New Focused Ion Beam (FIB) procured, delivered, and installed
- Contracts for FIB / EPMA Shielding and Confinement, as well as SSPA Shielded Transfer Cell / Glovebox and Hood have been placed.
- New Transmission Electron Microscope (TEM) procured and delivered.
- Conceptual design of Mechanical Properties Cell complete.

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IMCL Challenges

- **System testing on "inherited" SSPA revealed that manipulator ports were not leak tight – new manipulator seal tubes are needed and the SSPA will need to be modified.**
  - Engineering evaluation has determined how to modify the SSPA (in-situ) and the needed diameter of the seal tubes
  - Contract for purchase of new seal tubes has been sent to vendor for approval.
  - Seal tube delivery will take approximately 4-6 months.
- **Design/Fab of Shielding and Confinement systems are projected to take 44 weeks based on schedule from vendor – substantially longer than originally planned.**
  - Equipment / Confinement interface represents innovative approach to these systems – want to maximize equipment outside of confinement for ease of ops/maintenance while providing robust confinement of samples.
- **Mechanical Properties Test Cell conceptual design includes capabilities and provisions consistent with NR input – however, design and fabrication costs are expected to exceed base planning estimates.**
  - A constructability review of the design is being conducted. If refined estimated cost exceeds planning, additional discussions will be needed ensure funding matches customer needs - NR needs (cask handling) currently seen as requiring resolution.
  - Lessons learned will be applied to SPL Mechanical Properties cell

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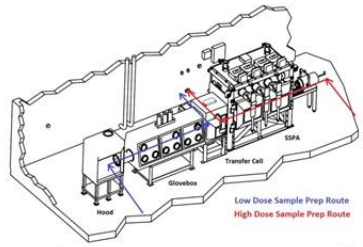
SSPA Plans

- Plans proceeding with boring out manipulator through tubes and purchase of new seal tubes and boot covers.
- Design of Shielded Transfer Cell (STC) and glovebox / hood system nearing completion. Fabrication expected to begin in March.
- Shielded Transfer Cell/glovebox line installation in FY15.



FY2014 | FY2015 | FY2016

Plan - Prepping Cold Samples ◆ Install Shielded Transfer Cell and Glovebox / Hood system with the SSPA ★ ★ Sample Prep Capability for low and high dose rate material

Readiness Activities – Calendar Year 2015

- Testing on surrogate samples to optimize remote operation of sample preparation tooling and new optical microscope continuing
- Complete line to be operational in early FY16

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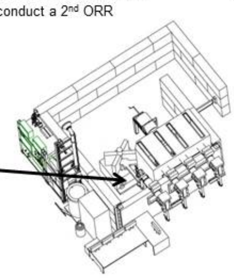
EPMA/FIB Plans

- Contract for Shielding and Confinement for EPMA and FIB signed and design work is proceeding.
  - Design implements unique approach with instruments outside of confinement to simplify vendor support and maintenance on microscopes.
  - Design emphasizes contamination confinement and radiological safety.
  - Second ORR required prior to operations with shielding and confinement for full rad operability – EPMA and FIB readiness activities will be combined for time / cost efficiency

FY2014 | FY2015 | FY2016

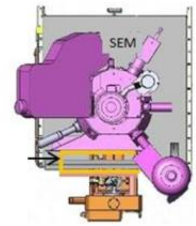
Plan - EPMA Installed in IMCL ◆ Fabricate S&C with unique interface. Bench-top work able to occur at IMCL | Install S&C in IMCL and conduct a 2nd ORR ★ EPMA with S&C Installed and Operational in IMCL

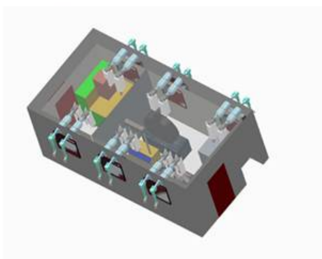
- In contrast to European designs, microscopy equipment will be placed outside of confinement.
- Adds complexity to design and fabrication of confinement / instrument interface.
- Results are better contamination control, decrease operations and maintenance costs long term.



Status of Other Equipment

- TEM delivered to INL in October 2014. Isolation room design on-going. Installation to be initiated in FY15.
- SEM shielding and confinement will be accelerated with new funding. Design and fabrication of shielded to leverage off of existing EPMA/FIB contract.



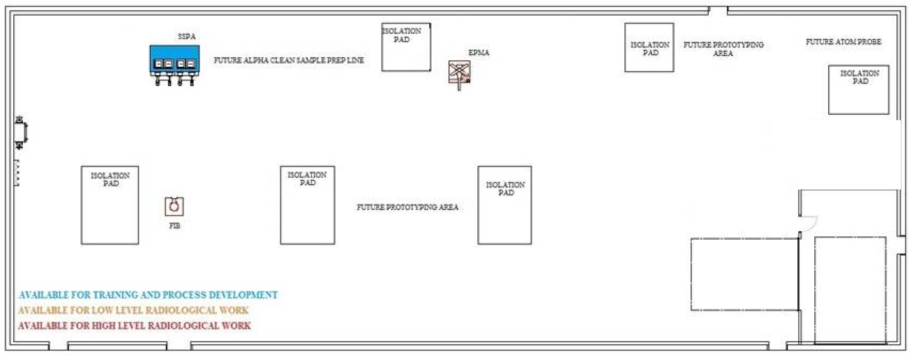


- Mechanical Properties Test Cell design on-going. Constructability review and cost estimate to determine size / capabilities of cell. Fabrication, procurement, installation, and operations expected in FY15 through FY17.
- Thermal Properties Cell design, procurement, installation, and operations expected in FY16 and FY17. Design efforts will be limited until NE programs complete instrument capability review (September 2015).

IMCL Equipment Installation Path Forward

- **FY 2015 –**
  - Complete readiness reviews to initiate bench-top (low rad/contamination levels) R&D activities on EPMA and FIB
- **FY 2016**
  - Complete installation and readiness reviews of shielded transfer cell, glovebox and hood attached to SSPA
  - Complete Installation and readiness reviews of shielded enclosures/confinement for FIB & EPMA to conduct R&D
  - Complete Installation and readiness reviews for TEM to initiate bench-top activities
- **FY 2017**
  - Complete installation and readiness reviews of shielded enclosures/confinement for thermal properties cell to conduct R&D
  - Complete installation and readiness reviews of shielded enclosures/confinement for SEM to conduct R&D
  - Complete installation and readiness reviews of shielded enclosures/confinement for Mechanical Properties Test Cell to conduct R&D

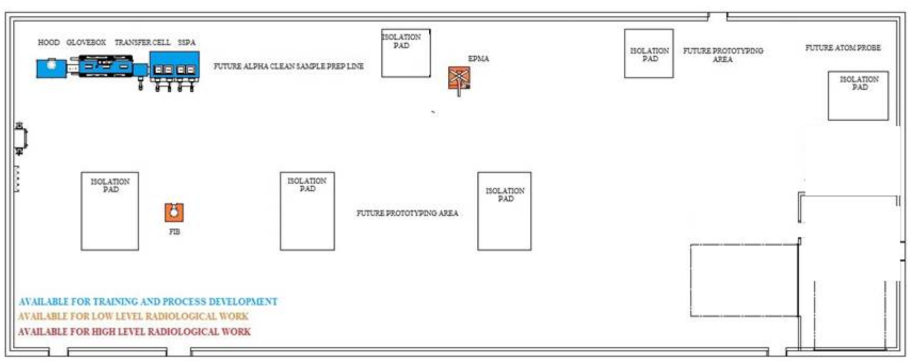
IMCL Floor Layout EOY FY14



IMCL Equipment
Installation Plan
29-Apr-15

IMCL Floor Layout EOY FY15

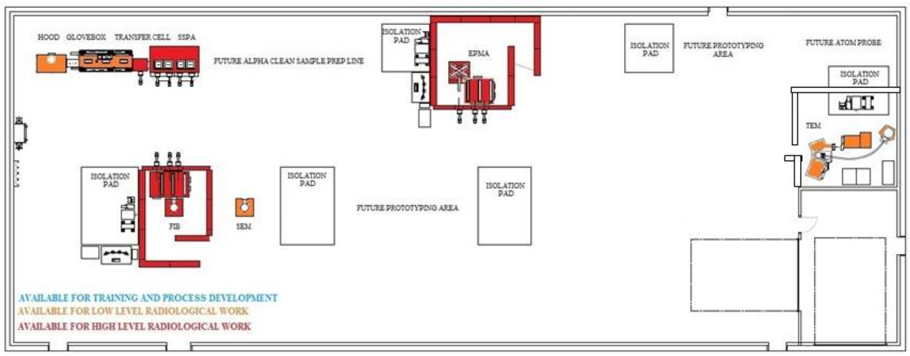
- Initiation of radiological work on FIB and EPMA



IMCL Equipment
Installation Plan
29-Apr-15

IMCL Floor Layout EOY FY16

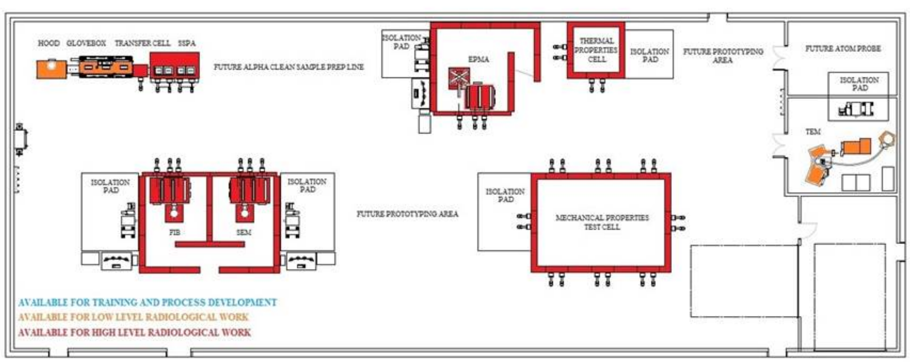
- Complete installation of shielded transfer cell, glovebox and hood attached to SSPA
- Installation of shielded enclosures/confinement for FIB and EPMA to conduct
- Installation of TEM



IMCL Equipment
Installation Plan
29-Apr-15

IMCL Floor Layout EOY FY17

- Installation of shielded enclosures/confinement for thermal properties cell to conduct R&D
- Installation of shielded enclosures/confinement for SEM to conduct R&D
- Installation of shielded enclosures/confinement for Mechanical Properties Test Cell to conduct R&D



IMCL Equipment
Installation Plan
29-Apr-15

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IMCL Capability Start-up Schedule

- Planned capabilities for IMCL:

Capability	Estimated Date Operational
Shielded Sample Preparation Area (SSPA)	January 2016
Focused Ion Beam (FIB)	May 2015
Electron Probe Micro-Analyzer (EPMA)	May 2015
Support Glovebox/Hood	January 2016
Shielding and Confinement for FIB	October 2016
Shielding and Confinement for EPMA	October 2016
Transmission Electron Microscope (TEM)	December 2015
Shielded Mechanical Properties Test Cell	November 2016
Scanning Electron Microscope (SEM) w/ Shielding and Confinement	September 2017
Thermal Properties Shielded Cell	September 2017

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Sample Prep Laboratory (SPL)

"Formally known as APIE"

- Since the approval of the original Advanced PIE (APIE) mission need statement in January 2011, the vision/scope of the facility mission has been refined to reflect updated R&D projections, funding realities, etc.
- The advanced PIE facility recommended in the earlier alternatives analysis was based on an expected significant increase in research budgets and subsequent program demand for throughput through the facility.
- The initial gap envisioned after the re-evaluation is smaller and more focused on the near-term advanced PIE needs.

Sample Prep Laboratory (SPL)

- A review of existing facilities shows that the advanced PIE drivers could be fulfilled by a combination of existing facilities/capabilities and a new facility to house additional sample preparation and dedicated mechanical property test equipment.
- Deployment of a smaller focused facility, strategically located next to other PIE facilities, with the inclusion of key missing capabilities will meet the objectives of the alternatives analysis and close the APIE gap.

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SPL Objectives

- A facility to support non-destructive and macro-destructive needs (HFEF) and nano-scale characterization (IMCL) would form the back-bone of advanced PIE capabilities.
- The remaining gap, consisting of improved sample preparation and handling along with mechanical property capability will be addressed by equipment and capabilities within a new facility.
- Efficient sample delivery of sufficient throughput between the three facilities (Hot Fuel Examination Facility, IMCL, SPL) will be a key component of the combined capability.

SPL Proposed Capabilities

Based on these objectives, pre-conceptual development has identified examples of new equipment/capabilities that may be housed in the new facility. These include:

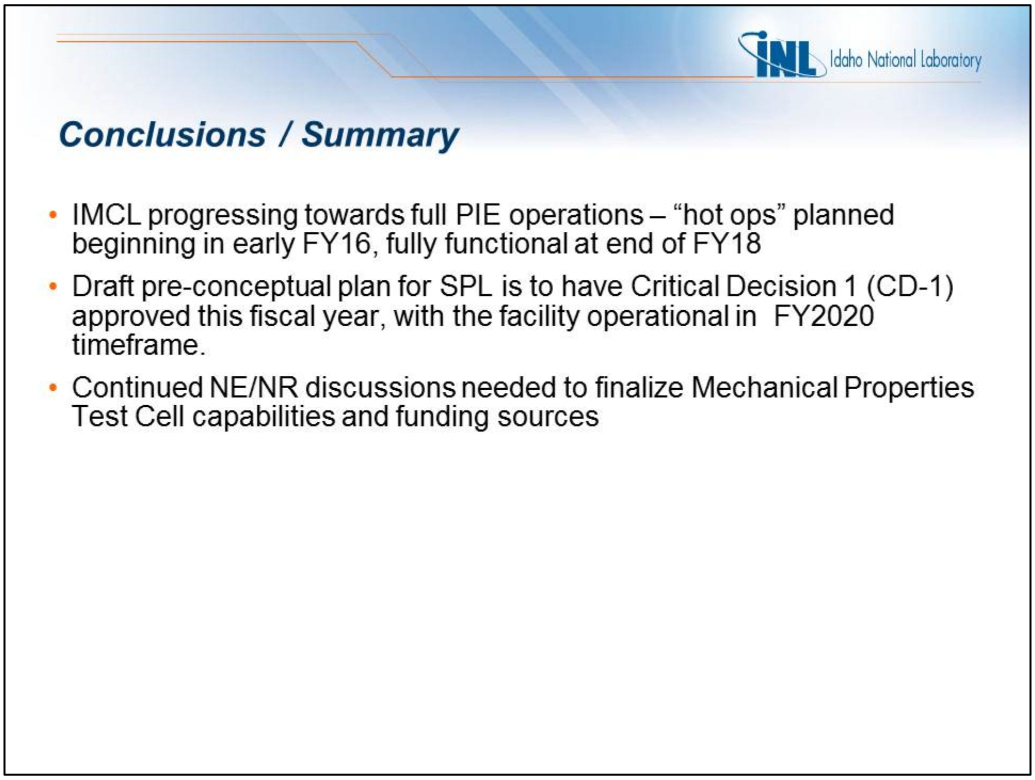
- Load frame and charpy testing machines.
- Micro-hardness tester to determine material properties.
- Scanning electron microscope for fracture surface analysis.
- Electric discharge machine (EDM) and sample preparation machinery (lathe/mill) for preparation of mechanical property specimens.

SPL Proposed Capabilities

- Hot cell for sample preparation of non-alpha bearing materials.
- Cask handling, with the capability to receive casks utilized across the complex.
- Appropriate space for program growth.

Technology improvements are regularly changing the way researchers study and research materials. The new facility is planned to be flexible and reconfigurable to adapt to emerging research needs.

Project Timeframe – 2018 design complete, 2020 construction complete/facility operational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Conclusions / Summary
IMCL progressing towards full PIE operations – "hot ops" planned beginning in early FY16, fully functional at end of FY18
Draft pre-conceptual plan for SPL is to have Critical Decision 1 (CD-1) approved this fiscal year, with the facility operational in FY2020 timeframe.
Continued NE/NR discussions needed to finalize Mechanical Properties Test Cell capabilities and funding sources