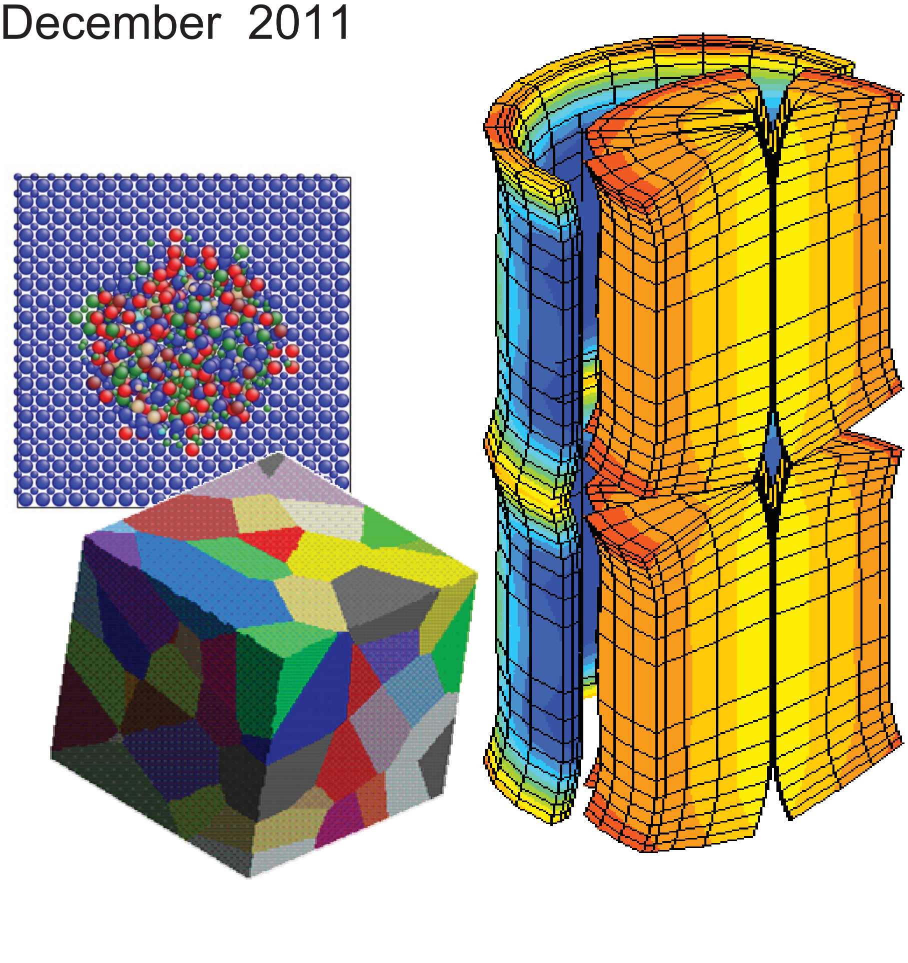
INL/EXT-11-23544

International Workshop on Characterization and PIE Needs for Fundamental Understanding of Fuels Performance and Safety

June 16-17, 2011
Paris, France

December 2011



Idaho National Laboratory

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Idaho National Laboratory
Idaho Falls, Idaho 83415

http://www.inl.gov

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EXECUTIVE SUMMARY

Global energy consumption is expected to increase dramatically in the next decades, driven by rising standards of living and growing population worldwide. The increased need for more energy will require enormous growth in energy generation capacity, more secure and diversified energy sources, and a successful strategy to tame greenhouse gas emissions. Among the various alternative energy strategies, building an energy infrastructure that capitalizes on nuclear energy may enable a secure and clean energy future worldwide.

Developing advanced nuclear fuel for light water reactors (LWRs) and new innovative fuels for future reactor designs will require an International collaborative effort. Characterization and postirradiation examination (PIE) capability is very important to advance nuclear energy as an option to meet global energy goals. Understanding the behavior of fuels and materials in a nuclear reactor environment is the limiting factor in nuclear plant safety, longevity, efficiency, and economics. State-of-the art characterization and PIE capabilities are required to transition into a “science-based” approach in the development of advanced fuels and materials, enabling a fundamental understanding of the behavior under irradiation.

The *International Workshop on Characterization and PIE Needs for Fundamental Understanding of Fuels Performance and Safety* was held June 16-17, 2011, in Paris, France. The Organization for Economic Co-operation and Development (OECD), Nuclear Energy Agency (NEA) Working Party on the Fuel Cycle (WPFC) sponsored the workshop to identify gaps in global capabilities that need to be filled to meet projected needs in the 21st century.

First and foremost, the workshop brought nine countries and associated international organizations, together in support of common needs for nuclear fuels and materials testing, characterization, PIE, and modeling capabilities. Finland, France, Germany, Republic of Korea, Russian Federation, Sweden, Switzerland, United Kingdom, United States of America, IAEA, and ITU (on behalf of European Union Joint Research Centers) discussed issues and opportunities for future technical advancements and collaborations.

Second, the presentations provided a base level of understanding of current international capabilities. Three main categories were covered: (1) status of facilities and near term plans, (2) PIE needs from fuels engineering and material science perspectives, and (3) novel PIE techniques being developed to meet the needs. The International presentations provided valuable data consistent with the outcome of the National Workshop held in March 2011.

Finally, the panel discussion on 21st century PIE capabilities, created a unified approach for future collaborations. In conclusion, (1) existing capabilities are not sufficient to meet the needs of a science-based approach, (2) safety issues and fuels behavior during abnormal conditions will receive more focus post-Fukushima; therefore we need to adopt our techniques to those issues, and (3) International collaboration is needed in the areas of codes and standards development for the new techniques.

The problems and needs discussed in the workshop are too big for any single country or institution to overcome. An International characterization and PIE capability development project is an ideal solution but that requires dealing with a number of obstacles in terms of international transport of radioactive materials, codes and standards, and regulatory requirements. However, if these obstacles can be overcome, the benefits would be tremendous in terms of advancing the performance and safety of nuclear energy worldwide. Please contact Lori Braase, lori.braase@inl.gov, or Kemal Pasamehmetoglu, kemal.pasamehmetoglu@inl.gov, for information regarding the workshop or this report.

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ACRONYMS

μ	micro
AES	Auger Electron Spectroscopy
AFM	Atomic Force Microscopy
AGR	Advanced Gas Reactor
AHTR	Advanced High Temperature Reactor
ANL	Argonne National Laboratory
ATR-NSUF	Advanced Test Reactor-National Scientific User Facility
BESAC	Basic Energy Sciences Advisory Committee
CEA	Commissariat à l'Energie Atomique
CO	Carbon Monoxide
DBTT	Ductile-Brittle Transition Temperature
DOE	U.S. Department of Energy
DPA	Displacements per atom
EDM	Electrical Discharge Machining
EDS	Energy Dispersive Spectroscopy
EELS	Electron Energy Loss Spectroscopy
EMI	electromagnetic interference
EPMA	Electron Probe Micro Analyzer
EPRI	Electric Power Research Institute
FCRD	Fuel Cycle Research and Development
FG	fission gases
FHR	Fluoride Salt-cooled High Temperature Reactor
FIB	Focused Ion Beam
FP	fission products
GCR	Gas Cooled Reactor
GNF	Global Nuclear Fuel
HR-TEM	High Resolution Transmission Electron Microscope
HTGR	High Temperature Gas Cooled Reactor
HTR	High Temperature Reactor
HTTR	High Temperature Test Reactor
IASCC	Irradiation Assisted Stress Corrosion Cracking
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
IMCL	Irradiated Materials Characterization Laboratory
INL	Idaho National Laboratory
IVEM	Intermediate Voltage Electron Microscope

LANL	Los Alamos National Laboratory
LECA-STAR	Postirradiation Examinations Laboratory at CEA-Cadarache
LLNL	Lawrence Livermore National Laboratory
LOCA	Loss-of-Coolant Accident
LWR	Light Water Reactor
M&S	Modeling and Simulation
MaRIE	Matter-Radiation Interactions in Extremes (LANL)
NDE	Non-destructive Examination
NE	Department of Energy Office of Nuclear Energy
NEA	Nuclear Energy Agency
NGNP	Next Generation Nuclear Plant
NMR	Nuclear Magnetic Resonance
NRA	Nuclear Reaction Analysis
NRC	Nuclear Regulatory Commission
NRF	Nuclear Resonance Fluorescence
OECD	Organization for Economic Co-operation and Development
OIM	Orientation Imaging Microscopy
ORNL	Oak Ridge National Laboratory
PBR	Pebble Bed Reactor
PDF	atomic pair distribution function
PIE	Postirradiation Examination
PMR	Prismatic Modular Reactor
PNNL	Pacific Northwest National Laboratory
R&D	Research and Development
SANS	Single Angle Neutron Scattering
SAXS	Single Angle X-ray Scattering
SCC	Stress Corrosion Cracking
SEM	Scanning Electron Microscope
SiC	Silicon Carbide
SIMS	Secondary Ion Mass Spectrometry
SNL	Sandia National Laboratory
STDM	Statistical Time Division Multiplexing
STEM	Scanning Transmission Electron Microscope
TAMU	Texas A&M University
TEM	Transmission Electron Microscope
TRISO	Tristructural-Isotopic Fuel

XAS	X-ray Absorption Spectroscopy
XPS	X-ray Photoelectron Spectroscopy
XRD	X-ray Diffraction

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International Workshop on Characterization and PIE Needs to Support Science-Based Development of Innovative Fuels

1. INTRODUCTION

The coupled challenges of a doubling of the world's energy needs by the year 2050 and increasing demands for "clean" energy sources that do not add more carbon dioxide and pollutants to the environment have resulted in increased worldwide attention to the possibilities of advanced nuclear energy as a long-term solution for a secure energy future. Increased interest in nuclear energy also triggered the search for next generation of nuclear fuel with enhanced performance and safety, and with reduced waste generation for the light water reactors that are currently used worldwide and that will continue to remain the preferred nuclear power technology for decades to come.

At the same time, other reactor technologies are also being developed to increase the use of nuclear energy beyond just electricity production (process heat applications). Small modular reactors are being considered because of their lower capital cost and their potential deployment to meet local energy demands. To enhance the utilization of natural resources and to minimize the amount of transuranic waste, fast reactors are likely to be the technology choice for the future. Many different innovative fuel types are being considered for these reactors and considerable fuels research is being conducted in various countries that are currently using nuclear energy or that are considering nuclear energy as part of their energy portfolio in the near future. Furthermore, after the events in the Fukushima plants in Japan, there is a renewed emphasis on understanding the fuel behavior during beyond-design basis events.

An internationally coordinated effort can accelerate the transition to state-of-the art material science for nuclear fuels and cladding materials

Fuel development (including the cladding) relies heavily on understanding the fuel performance under neutron irradiation. The major part of the research is devoted to irradiating samples of candidate fuel and cladding types in reactors. Developing an advanced characterization and postirradiation examination (PIE) capability is critical the fuels and materials research programs across the globe. Understanding the behavior of fuels and materials in a nuclear reactor environment is a limiting factor in nuclear plant safety, longevity, efficiency, and economics. State-of-the art characterization and PIE capabilities are required to transition into a "science-based" approach in the development of advanced fuels and materials, enabling a fundamental understanding of fuels and materials behavior under irradiation. In recent decades, considerable advances are made in instrumentation technologies as applied to material science. However, most of those advances are not translated into the nuclear energy field. Application of such technologies and methods to highly irradiated materials will provide considerable advances compared to today's PIE capabilities. Many countries are increasing their PIE capabilities and with an internationally coordinated effort, the transition to state-of-the art material science for nuclear fuels and cladding materials, can be accelerated to benefit everyone.

1.1 Background

The traditional approach to fuel development is generally empirical and relies mainly on measurements of engineering scale performance parameters before and after irradiation testing. This empirical approach is time consuming and expensive. Furthermore, the engineering-scale parameters that are measured provide an integral view of multiple, strongly interactive phenomena that occur at lower-length and time scales.

Phenomenological understanding of irradiation behavior is not directly possible through traditional PIE efforts because fuel development requires multiple iterations and does not readily enable the scientists and engineers to design fuels and materials for the desired performance. A science-based fuel development approach aimed at a fundamental understanding of fuel and cladding behavior under irradiation requires measurements and understanding at the lower-length and time scales. Such measurement capabilities for high-dose materials and irradiated fuels are being developed internationally in multiple locations.

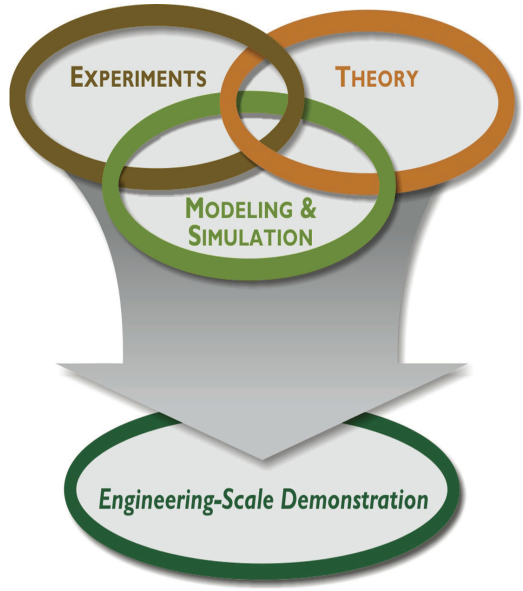

Figure 1. Major Elements of a Goal Oriented Science-Based Approach (Presentation 8).

The "Goal Oriented Science-Based" approach, depicted in Figure 1, combines theory, experimentation, and high-performance modeling and simulation to develop the fundamental understanding that will lead to new technologies. Advanced modeling and simulation tools will be used in conjunction with smaller-scale, phenomenon-specific experiments informed by theory to reduce the need for large, expensive integrated experiments. Insights gained by advanced modeling and simulation can lead to new theoretical understanding and, in turn, can improve models and experimental design. The different length and time scales relevant to fuels and materials behavior under irradiation is depicted in Figure 2. The ellipse with yellow shade indicates that the traditional PIE domain at the engineering-scale is presently being extended into the mesoscale region with measurements at the microstructural level becoming more and more routine.

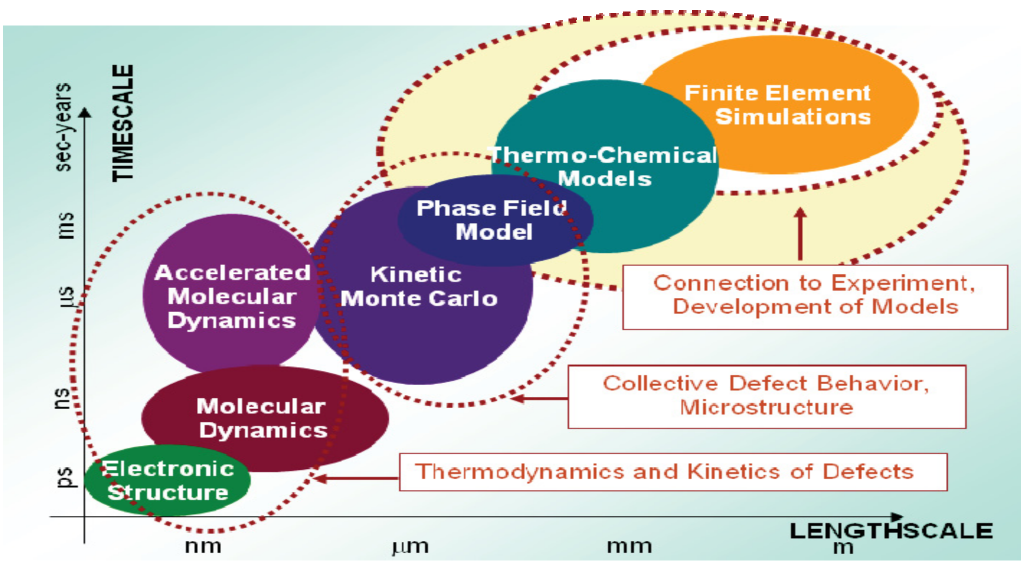

Figure 2. The length and time scales relevant to understanding fuels and materials under irradiation (Presentation #1).

The first step to achieve a state-of-the art characterization and PIE infrastructure is to adapt currently available instruments and scientific techniques for use with highly radioactive fuels and material samples.

The objective is to start characterizing radioactive samples at the nano-to-micro scale resolutions. Some of these instruments, when applied to radioactive materials characterization, require special facilities with very strict environmental control.

International collaborations are extremely valuable to characterization of radioactive samples at the nano-to-micro scale resolutions.

The second step is to design and develop instruments that currently do not exist but are required to measure properties at various length and time scales in order to support a complete fundamental understanding of radioactive materials behavior. These instruments would be valuable in understanding separate effect phenomenology and supporting the multi-physics, multi-scale, predictive fuel performance modeling efforts. These sophisticated capabilities will require new facilities with specialized environmental control and integration among multiple techniques.

International collaborations in terms of sharing experiences, technique development, samples, and benchmark results will be extremely valuable in achieving the objective. This was the primary motivation of this workshop.

1.2 International Workshop Overview

The *International Workshop on Characterization and PIE Needs for Fundamental Understanding of Fuels Performance and Safety* was held at OECD-NEA on June 16, 2011, in Paris, France.

The objectives of the workshop were

- To review the existing characterization and PIE capabilities and the novel techniques that are being developed at the international facilities
- To identify the gaps to meet the needs of 21st century characterization and PIE capabilities.

The scope of the workshop included a series of presentations by experts from various organizations on the status of existing facilities, capabilities and near-term plans. The presentations covered related activities in IAEA, Europe, South Korea and the U.S. It was not possible to cover all the existing and planning capabilities world-wide, but a considerable cross-section was represented in the workshop.

The second topic of discussion was the characterization and PIE needs for 21st century fuel development efforts. The topics were addressed through invited presentations by international experts, covering modeling, fuel design, and safety perspectives.

A third topic covered during the workshop was focused on new techniques that are being developed at various institutions, such as those being used in the French LECA-STAR facility, as well as beam-lines, X-Ray absorption, X-ray microtomography, and laser-based acoustic techniques.

Finally, a panel discussion was organized to discuss the integration of testing, characterization, and PIE with modeling and simulation. Safety was one of the topics relative to the Fukushima Daiichi accident in Japan earlier this year. One of the issues is whether we currently have the right PIE equipment to determine fuel behavior when cladding fails or how steams reacts with the cladding and the failed fuel. Other topics covered by the panel discussion include thermodynamics, thermal conductivity, irradiation conditions, microstructure, geometry, and properties.

The workshop was attended by representatives from nine countries (Finland, France, Germany, Republic of Korea, Russian Federation, Sweden, Switzerland, United Kingdom and United States of America), and two International Organizations (International Atomic Energy Agency and Institute for Transuranium Elements). The workshop participants are listed in Appendix A.

Unfortunately, the experts from Japan were not able to join the workshop because of the disastrous earthquake and tsunami in Japan, followed by the unfortunate events in the Fukushima power plants. Their expertise and contributions to the discussions were surely missed.

2. REVIEW OF INTERNATIONAL PIE CAPABILITIES AND PLANS

A summary of the presentations related to PIE capabilities and future plans is provided in this section. The relevant presentations are included in Appendix B.

2.1 IAEA PIE Database

Dr. Uddharan Basak (IAEA) reviewed the agency's activities related to the PIE database (HOTLAB PIE Database). The database was created in 1990 and published in 1996. It was converted to an electronic database in 2003 and modifications and updates were last added in 2010. It covers 42 facilities in 22 countries and information for 19 casks. The database contains general information and cell characteristics, acceptance criteria for irradiated components, available PIE technology, refabrication and instrumentation capabilities, storage and conditioning capabilities, and technical and licensing data for casks. The database appears to be a very valuable source for the hot-cell based PIE capabilities. However, PIE capabilities that may not require standard hot-cells (shielded boxes, small-cells with removable walls, etc.) that may be more appropriate with the modern instruments and that require analyses of very small samples are not included in the database. Additional information can be accessed at http://www-nfcis.iaea.org/PIE/PIEMain.asp

2.2 PIE Capabilities in KAERI

Dr. ByoungOon Lee (KAERI) provided an overview of metallic fuel development activities in Republic of Korea with emphasis on PIE capabilities. The presentation included the overview of the IMEF (Figure 3) where the PIE capabilities reside. PIEF (Figure 4) also is used for testing and evaluating the performance and integrity of fuel discharged from the reactor. IMEF includes the necessary nondestructive and destructive examination capabilities with state-of-the art equipment. At present, the emphasis of this facility is on meeting the needs of traditional PIE, but certainly offers opportunities for implementing new capabilities as well.

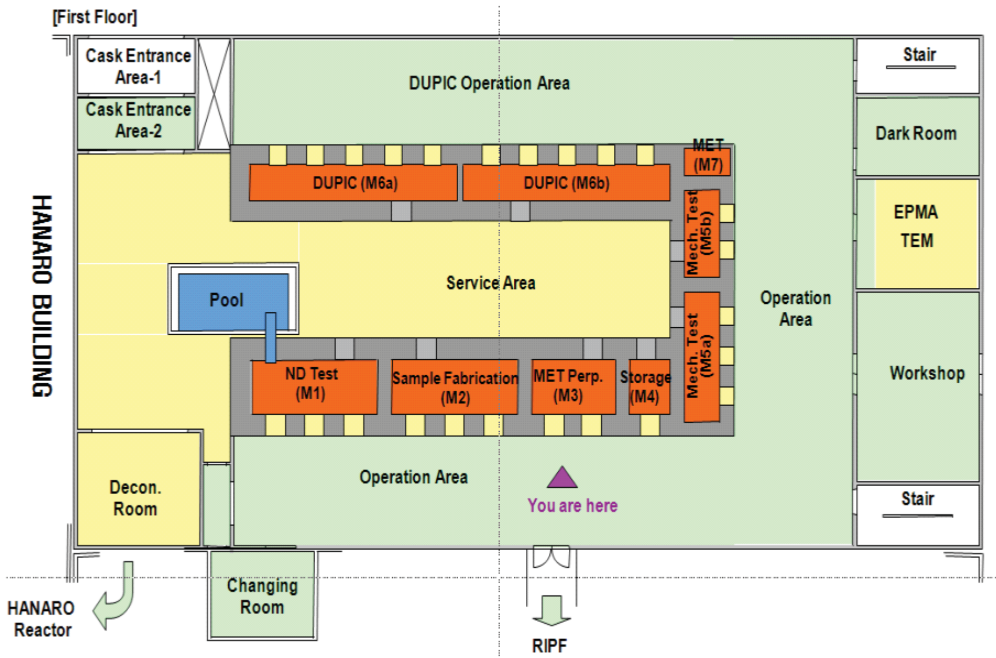

Figure 3. First floor plan view in IMEF (Presentation #4).

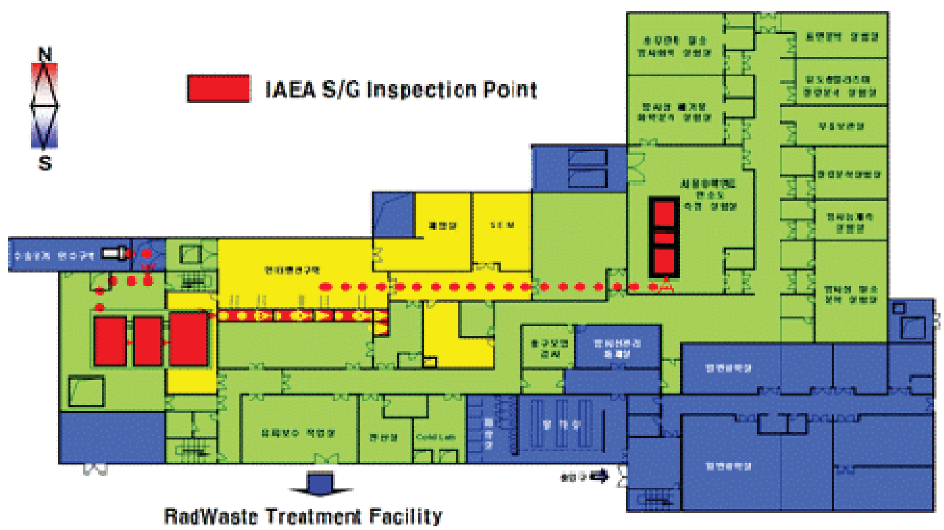

Figure 4. First floor plan view in PIEF (Presentation #4).

2.3 Examples of PIE Capabilities in Europe

A general overview of the CEA LECA-STAR (Figure 5) facility in France was provided by Dr. Jerome Lamontagne and Dr. Francois Sudreau. Dr. Lamontagne provided an overview of equipment and capability, such as:

- SEM - morphological analyses
- EPMA – quantitative method of non-destructive elemental analysis
- SIMS – isotopic and gas analysis
- XRD – crystalline analyses
- SEM-FIB - 3D imaging of microstructure and preparation of thin samples
- EBSD - interaction between a crystal and the primary electrons
- TEM - microstructure analysis at a nanometric scale
- MARS - multipurpose beamline.

Dr. Sudreau's presentation (see Appendix B) included a specific example of in cell testing and state-of-the art measurement techniques for fission gas release behavior in irradiated fuels.

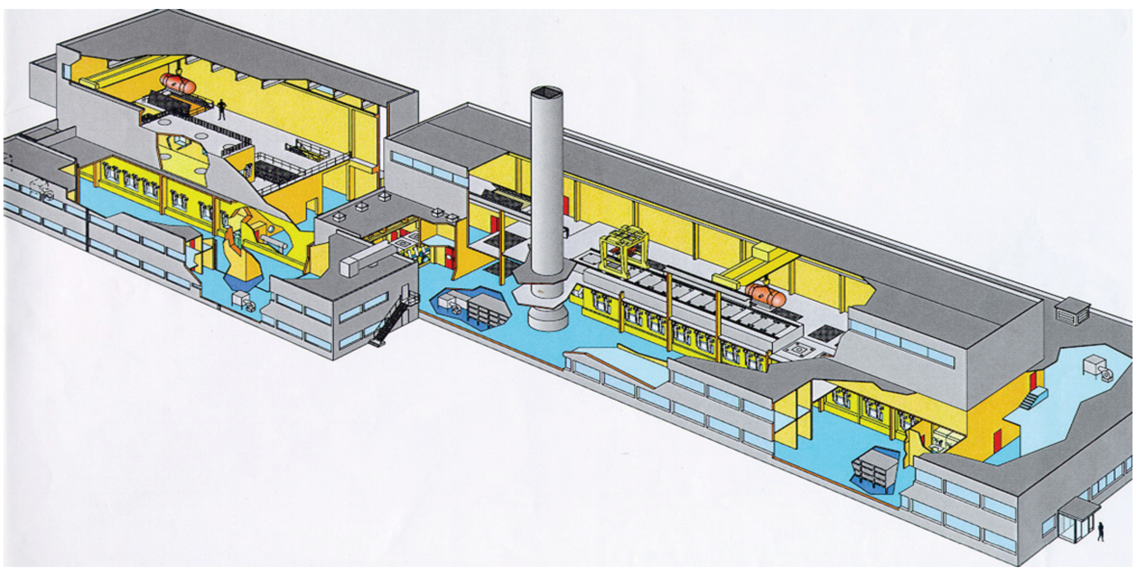

Figure 5. Layout of LECA-STAR facility (Presentation #13).

Dr. Joe Summers (ITU) presented the TEM Facility development, which is a new addition to their capabilities. The presentation also summarized the activities associated with the refurbishment and replacement of aging equipment in European facilities (CEA, JRC-ITU, SCK-CEN, and NRG) including the new TEM capabilities. Figure 6 is a picture of the Tecnai G^2 F20 XT, which is remote controlled and adapted for nuclear materials.

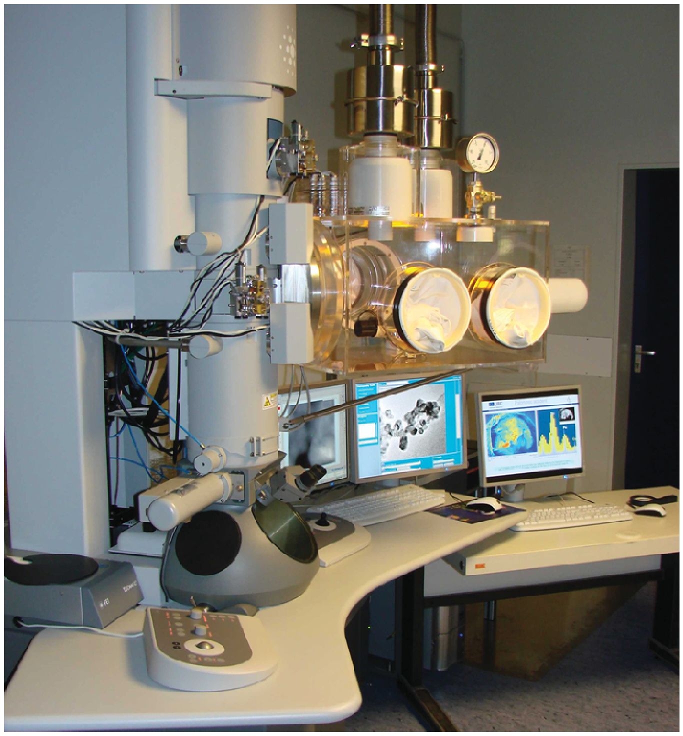

Figure 6. Tecnai™ G^2 F20 XT (Presentation #3).

2.4 Summary of PIE Capabilities in the U.S.

The U.S. presentation was provided by Dr. Dennis Miotla (U.S. DOE). The presentation provided a short summary of existing capabilities and future facilities with state-of-the art equipment. Dr Miotla emphasized DOE's plan to invest in facilities and capabilities that are adaptable to changing technologies, sustainable, and most importantly, accessible to researchers worldwide. Figure 7 show a drawing of the Irradiated Materials Characterization Laboratory (IMCL) at the Idaho National Laboratory (INL). This facility is currently under construction and should be complete by the end of 2012. It will house PIE equipment in shielded cells designed to examine radioactive fuel and materials samples.

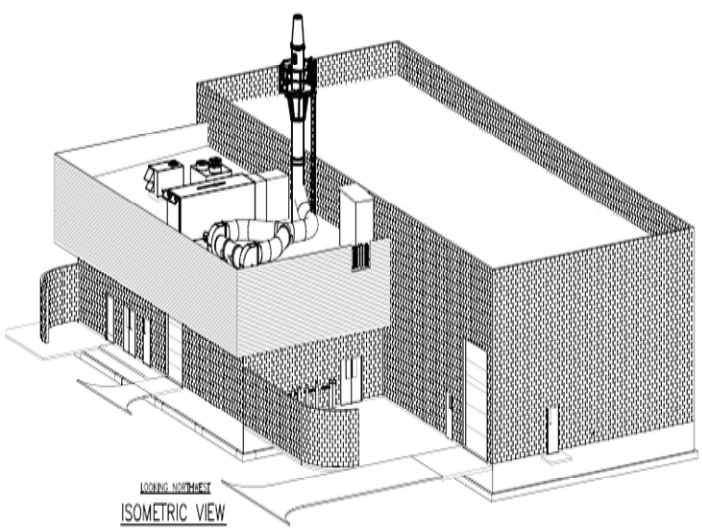

Figure 7. Irradiated Materials Characterization Laboratory (IMCL) (Presentation #5)

3. 21st CENTURY PIE NEEDS

In this session, three presentations were provided to outline the PIE needs from a modeling and simulation, fuel design, and nuclear engineering perspectives. All the related presentations are included in Appendix B.

3.1 Fuel Performance Codes: Characterization and PIE needs

Dr. Vincent Bouineau (CEA) presented performance code PIE needs for physical properties and behavior laws for code improvement and validation. Additional points of emphasis include:

- Analytical experiments on simulated fuels to understand basic physio-chemicals behavior (see Figure 8 for helium implanted UO_2 and NRA characterization)
- Analytical irradiation; some characteristics are fixed or adjusted during the irradiation and must be known also as function of radial (and axial) position.
- Integral or semi-integral irradiation
  - Characteristics needed as function of radial and axial position.
  - In the worse case after the fabrication and during PIE and in the best case all along the irradiation.

When modeling fuel with minor actinides, improvements MUST BE COMPLETED with

- More accuracy on fabrication data and irradiation conditions
- Analytical experiments
- More instrumentation in core, especially in MTR (also in prototype?)
- More characterizations in hot cell

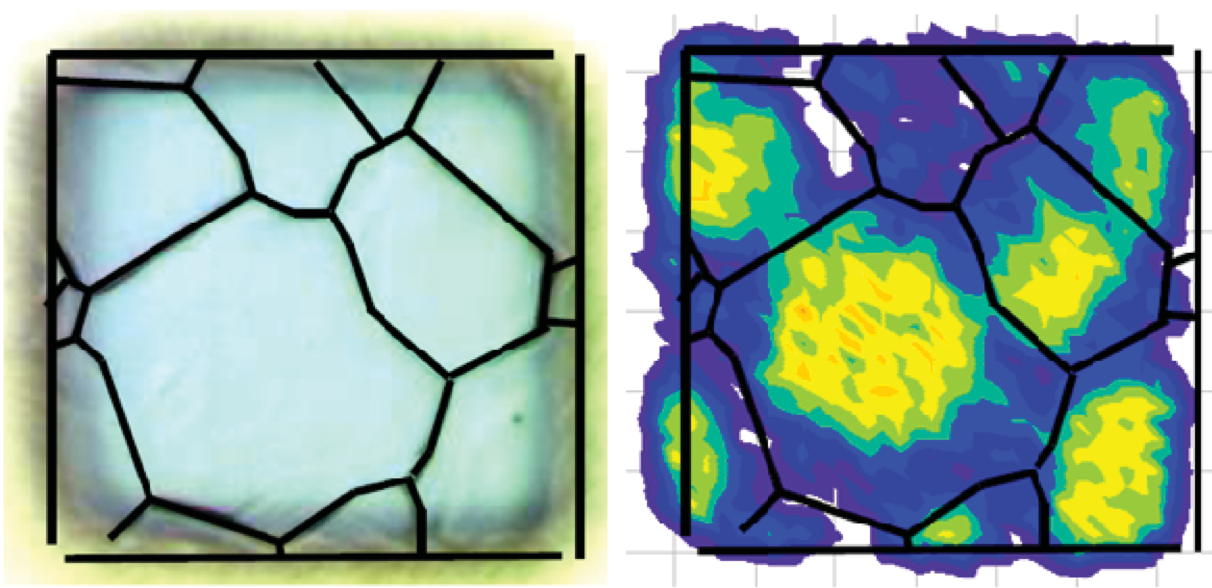

Figure 8. He implanted UO_2 and NRA characterization (Presentation #6). G. Martin, P. Garcia, C. Sabathier, G. Carlot, T. Sauvage, P. Desgardin, C. Raepsaet, H. Khodja , Nuclear Instruments and Methods in Physics Research Section B 268, 2133 (2010)

3.2 Atomistic Modeling: Characterization and PIE needs

Dr. Michel Freyss (CEA) presented atomistic modeling techniques (first-principles calculations, molecular dynamics with empirical potentials, cluster dynamics) and the corresponding characterization and PIE needs. He emphasized that most of the material data to validate the atomistic scale models, especially for innovative fuels such as mixed oxides and carbides, do not exist and are difficult to obtain. TEM, XAS, diffusion experiments, etc., are relevant techniques (see Table 2). The needs include material data (local crystal structure, electronic structure, chemical order, elastic constants, etc.), activation energies for atom diffusion, self-diffusion, and fission product diffusion coefficients, radiation damage, etc. Figure 9 is a representation of point defects and fission products in UO_2 as modeled using first-principles calculations.

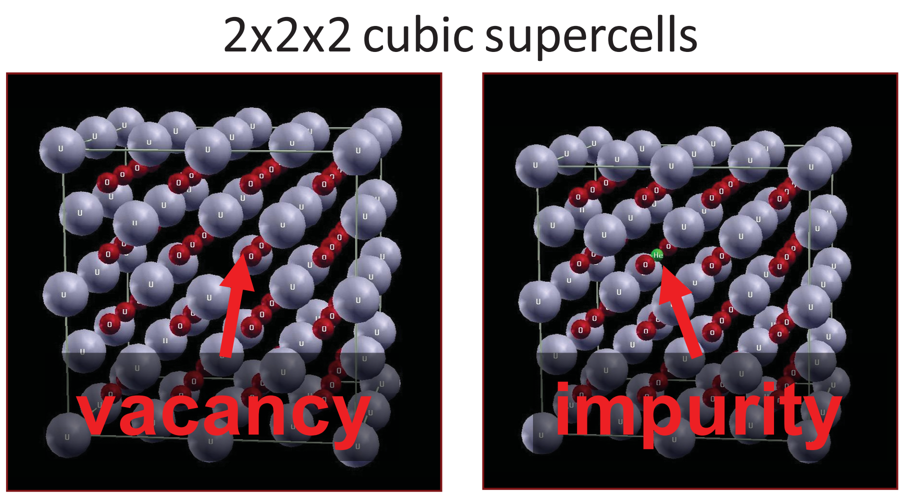

Figure 9. Point defects and fission products in UO_2 (Presentation #7).

3.3 Fuel Design and Safety Testing: PIE Needs

Dr. Steve Hayes (INL) presented the characterization needs from the fuel design and safety perspective. Standard PIE measurements are essential when using equipment with higher accuracy. However, those need to be supplemented by micro- to nano-scale characterization techniques to reach a fundamental understanding. This is primarily driven by the length and cost of licensing a new fuel. The limited access to certain facilities (e.g. fast test reactors) also requires a better understanding of the fuel behavior because only a few prototypic tests can be conducted during the development phase. Figure 10 is a picture of the in-cell Scanning Thermal Diffusivity Measurement (STDM) equipment at the INL.

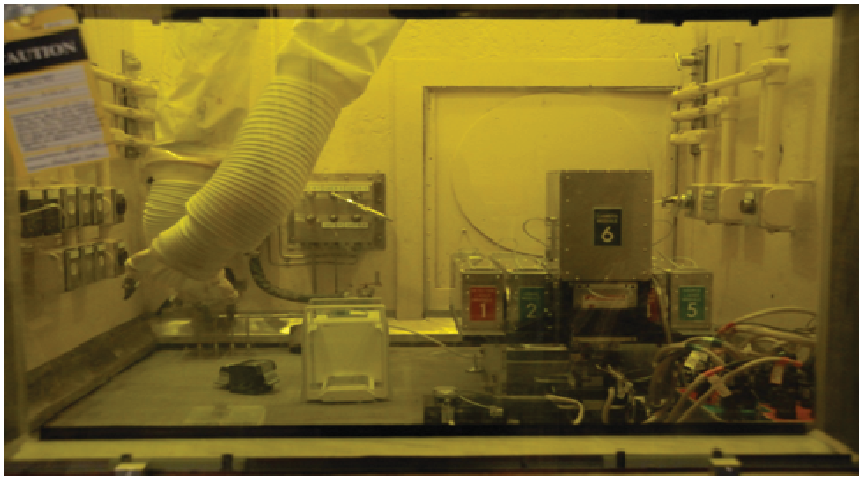

Figure 10. Scanning Thermal Diffusivity Measurement (STDM) technique (Presentation #8).

4. EXAMPLES OF NOVEL MEASUREMENT TECHNIQUES

A few examples of new techniques are discussed in this session. All the associated presentations are included in Appendix B.

4.1 Beam Line Measurements

Dr. Manuel Pouchon (PSI) presented an overview of the beam line facilities available for materials research. These include X-ray beam lines with synchrotron light sources and free electron lasers (FEL) as shown in Figure 11. Particle beams (neutrons and ion beams) were also discussed. The techniques provide valuable data at lower length and time scales and are particularly beneficial for data in support of atomistic models. However, access to the beam lines is limited and requires a long planning time. Furthermore, the use of irradiated fuel samples is currently not possible in most available beam lines.

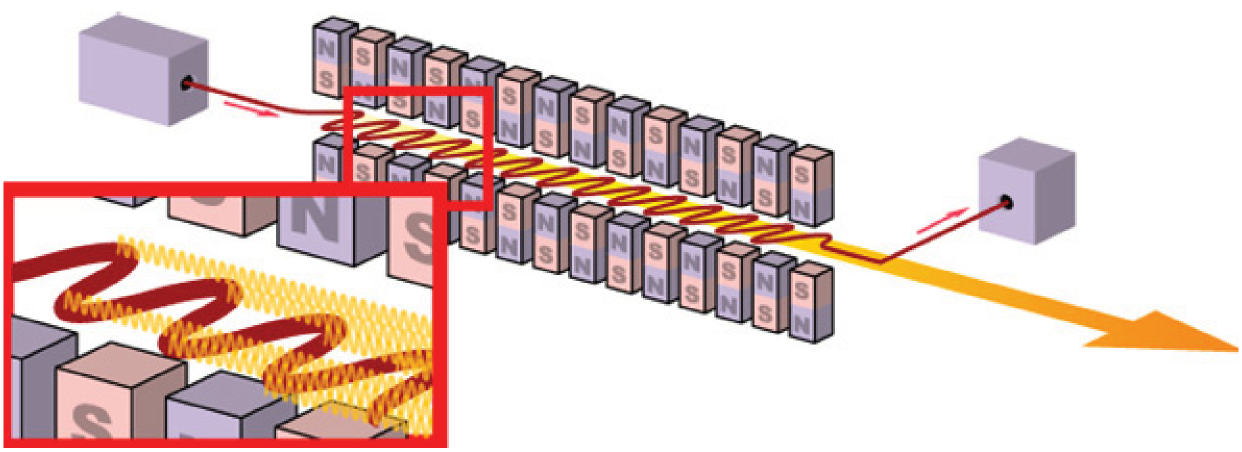

Figure 11. X-Ray Free Electron Laser (FEL) (Presentation #9).

4.2 Laser-Based Measurement Techniques

Dr Rory Kennedy (INL) presented the development of the laser-based measurement techniques. The development includes measurements (at micron-scale) for thermal diffusivity and thermal conductivity, average elastic constants, ultrasonic attenuation and microstructure, and dispersion relations. Because it can be remotely operated, this technique is amenable for in-pile, real time applications. Dr. Kennedy provided some examples of data being collected and also the development path for future applications. Figure 12shows the schematic for the Combined Laser Ultrasonics and Resonance Ultrasound Spectroscopy.

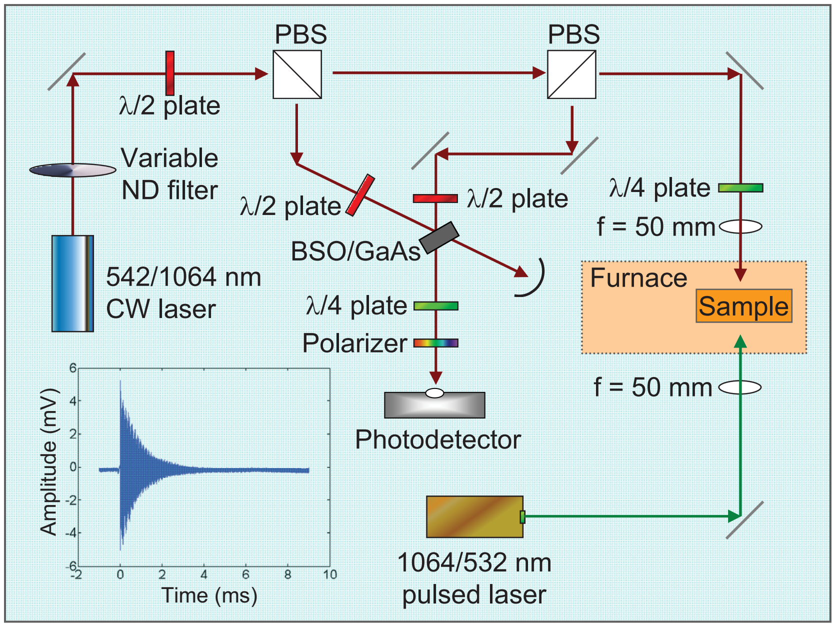

Figure 12. Combining Laser Ultrasonics and Resonance Ultrasound Spectroscopy (Presentation #10).

4.3 X-Ray Micro Tomography

Dr. Lance Snead (ORNL) presented the application of X-Ray microtomography technique to TRISO fuel development and graphite studies. Currently, the resolution is 1.4 µm (see Figure 13) but improvements are underway to obtain sub-micron resolution with a potential for 70 nm resolution. Also, the development at ORNL continues to apply this technique to samples with higher radiation dose.

Figure 13. Actual imaging resolution of current system surrogate TRISO particle at 1.4 µm resolution (Presentation #11).

4.4 Density Measurements with X-Ray Absorption

Dr. Somers (ITU) presented the x-ray absorption technique for local density measurements on nuclear materials. Figure 14 shows the X-Ray Powder Diffractometer schematic. The advantages of using X-Ray absorption as a complementary alternative to immersion techniques are listed below.

- Non destructive method, a fuel disk with parallel sides is required
- Not affected by temperature and pressure fluctuations in the hot cell
- Radial total porosity/density profiles on fuel pellets can be measured
- Easily repeatable and fast measurements

However, the technique has some disadvantages, such as:

- Fuel cracks must be avoided (similar to diffusivity measurements with laser flash)
- Difficult calibration (solid fission products cannot be considered, only empirical corrections)

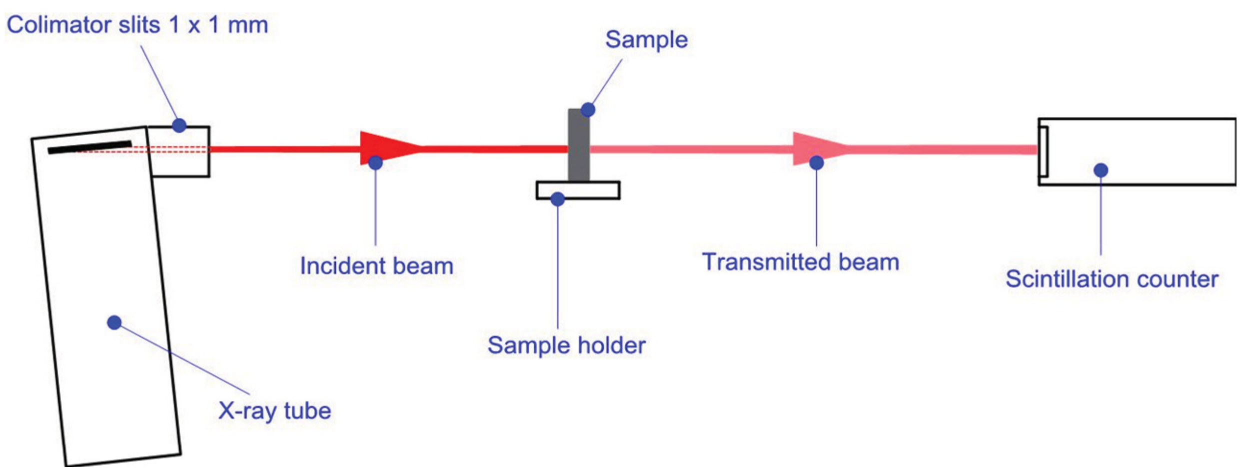

Figure 14. X-Ray Powder Diffractometer (Seifert XRD 3000 PTS) (Presentation #14).

5. GROUP DISCUSSION ON MODELING NEEDS

Due to the strong international support for modeling, the PIE and characterization needs were grouped to support fuel performance modeling and simulations. It was recommended that the modeling needs be prioritized by the areas that give us the biggest advantage (bang for the buck). An integrated approach should be used to close down on modeling needs quickly. Table 1 and Table 2 summarize the results of the discussion. Table 1 includes both the currently available techniques and the future possibilities to close the data gaps. For future capabilities, instruments and techniques are listed but those are not necessarily available inside a suitable facility at present.

Table 1. Needs for Performance Codes

Parameter	Device For Measurement on Irradiated Fuel
Composition	
Fuel composition: - U, Pu, MA - Isotopic composition - Fission products, impurities	- EPMA - EPMA+ SIMS + isotopic dissolution + mass spectroscopy - ICPMS - PIXE on active samples - X-ray fluorescence and absorption spectroscopy
Gases (helium and fission gases)	- EPMA + SIMS - TEM - EBSD - NRA - Laser ablation - X-ray fluorescence and absorption spectroscopy
Stoichiometry (O/M)	- EPMA - SIMS - Electrochemical Cell
Chemical form of each component inside the fuel (single or multiple phases)	- XRD and micro focus XRD - TEM - EXAFS - SIMS + EPMA
Interaction with other components (especially cladding)	- EPMA, SEM, TEM - SIMS - Beam line (some beam lines allow samples with MAs but irradiated fuel samples are not easily permissible)
Clad wastage	
Gas composition and pressure in the gap	- Puncturing + spectrometry analysis
Microstructure	
Density	- Tomography - Tomography 3D - Immersion
Porosity (open, closed, shape of porosity)	- SEM-FIB - Micro beam XR tomography may be a possibility
Grain size, cracks	- SEM-FIB - EBSD - Nano-indentation for cracks is a question
Columnar grain diameter	- Metallography
Bubbles interconnection	- SEM-FIB
Geometry	
Fissile column length,	- Gamma spectrometry et X-Ray
Gap closure and pellet/clad contact	- Diametral Metrology - Neutron Radiography with high resolution
Pellet diameter	- Metallography
Inner/outer clad diameter	- Diametral metrology, Eddy Current, US methods to be developed
Fuel central hole	- X-Ray, gamma spectrometry, metallography

Parameter	Device For Measurement on Irradiated Fuel
Properties	
Thermal diffusivity	- Laser flash
Thermal diffusivity	- Calorimeter
Thermal expansion	
Melting point	
Eutectics	
Young modulus	- Nano indentation
Creep	- Indentation

Table 2. Needs for Atomistic Codes

Modelling Type Parameter	Device For Measurement on Irradiated Fuel
Ab initio	
Material properties: composition, crystal structure, chemical order, elastic constants, phonon spectra, electronic structure, thermodynamic data, etc.	- XRD, XAS, NMR, Raman, PES (XPS and UPS)
Activation energies of diffusion (self-diffusion and gas diffusion)	- PAS, TEM
Empirical potentials	
Material properties	Same as ab initio
Material thermo-mechanical properties	Same as fuel performance code and ab initio
Radiation damage, microstructure evolution	- SEM-FIB, TEM, PAS, EBSD
Fission gas and helium bubbles	- SAX, TEM, NRA
Cluster dynamics	
Atomic diffusion coefficients	- TDS, SIMS
Size and density of fission gas and helium bubbles	- SAX, TEM

6. GROUP DISCUSSION ON SAFETY NEEDS

The PIE workshop took place shortly after the Fukushima nuclear plant accidents in Japan. Therefore it was decided to include safety needs as part of the workshop discussions. The following are the important conclusions and highlights that resulted from that discussion.

The current infrastructure is not sufficient to answer all the questions we face in case of considerable fuel and cladding damage. The primary questions are

- How does the cladding fail (rupture, melting, etc.)?
- What happens to fuel when cladding fails?
- How does steam react with cladding and fuel?

In conclusion, the current testing, characterization and PIE capabilities are not sufficient to fully answer the questions.

During the accidents, the phenomena were very complex and with thermodynamic non-equilibrium phases. The ability to collect and analyze the data during testing is important because it is hard to deconvolute the data after the testing to understand what is happening. Instrumented in-pile transient testing is very valuable up to and beyond failure, but the information that can be gathered is limited and such test reactors are not readily available for destructive testing. The discussion resulted in a number of strong recommendations.

1. *Hot-cell furnace tests combined with characterization and PIE to understand the detailed behavior under accident conditions.*

2. *Standard characterization and PIE equipment are not totally adequate for severe accident testing (temperature limitations, etc.). New measurement techniques should be explored, especially to measure chemical forms of the products.*

3. *Small size clad-fuel in pile experiments would be useful to gain fundamental understanding of local phenomena.*

4. *Additional properties data on irradiated fuel samples (emphasis on accident conditions)*

Since this type of testing is expensive, a strong international collaborative effort is essential to fully understand the fuel behavior under severe accident conditions. Some capabilities exist (e.g. Studvik, VERDON in STAR hot cells at CEA), some capabilities are being developed (e.g. STAR hot cells at CEA), some capabilities for continuous interrogation and characterizations are being considered (MaRIE at LANL) but much more is needed, along with some fundamental thermo-chemistry data relevant to accident conditions. This is a high-temperature thermodynamics problem with coupled fuel and cladding behavior.

The OECD/NEA Expert Group should consider adding severe accident testing to their charter as a means of starting the international collaborative effort. The first step would be a review of existing capabilities.

As a follow on step, the design of a test facility could be another international collaboration effort. The design would include the size and scope of testing, data needs, data collection methods, design of new instruments and techniques for data collection, data storage and analyses, etc. The involvement of safety authorities (e.g. NRC) would also be very valuable.

In conclusion, there is opportunity to look into a test facility design as an integral international capability to do testing and characterization simultaneously, in one place to understand the data.

7. HIGHLIGHTS AND MAJOR CONCLUSIONS OF THE WORKSHOP

The workshop confirmed that there is considerable infrastructure in place and new capabilities are being developed around the world. However, to fully-support the science-based development of fuels and cladding, and to understand their behavior under accident conditions, all the needed capabilities do not exist. It is not likely or desirable to have all of these capabilities in one location. An international collaboration where a set of capabilities is fully connected providing a collective characterization and PIE laboratory spread across the globe is a very strong possibility. Also, running new, large-scale experiments are difficult and expensive due to facility limitations (e.g. limited number of fast test reactors). Therefore, the information gained from a given experiment must be maximized using collective resources. The following are the major elements of such an international collaboration.

7.1 International PIE Project

- An international working group or an expert group can be formed to create and coordinate an International characterization and PIE project. The Innovative Fuels Expert Group can function during the early planning phases but a larger and more formal group will likely be needed for implementation.

- The group must provide a joint definition of the data requirements to lead towards a pure science-based predictive modeling approach, which in turn will define the types of characterization tests required and the necessary measurement techniques. This will also define the measurement technology shortfalls and the need for additional technique and equipment development work.
- The characterization and PIE needs may be different for the different phases of the fuel development process. It would be beneficial to define the needs as a function of technology readiness level and decision points during the development process, all the way up to licensing. Statistical issues become important as we approach the licensing phase.
- The licensing approach can be adapted to the new paradigm of science-based approach. However, this requires collaborations among the licensing authorities, scientists, and engineers in the early stages.
- Codes and standards vary between countries. Efforts to synchronize the codes and standards would be very beneficial.
- Funding limitations and expertise prevent doing all the work in one location. Resource sharing, especially for expertise and field experience, is important to success.
- Special sub-projects and teams can be established to address high priority needs (e.g. helium measurements in used fuel, full length tomography, etc.)

7.2 Testing Considerations

- Maximizing the information gained from a given irradiation testing is important by sharing samples and utilizing the collective resources.
- Transient testing capability, experiments, and data from such tests are limited. Strong collaborations on transient testing of fresh and irradiated fuels are needed.
- Combined testing and characterization facilities that can provide fundamental thermodynamic data to examine fuels and cladding under severe accident conditions do not exist.

7.3 Modeling and Simulation

- Advanced modeling and simulation is a powerful tool for fuel development. However, all the necessary data for model development and validation do not exist, especially at lower length and time scales. The ongoing strong international collaboration on modeling and simulation can be a driver for similar collaboration on data collection.
- Benchmarks among codes and models are important even when we compare them to data, because we do not measure everything in experiments. Code-to-code and model-to-model benchmarks allow for comparison of many more parameters. Favorable comparisons increase confidence in predictions.
- Standard benchmarks for codes developed in multiple institutions would be very beneficial.

7.4 Transportation and Sample Sharing

- Global transportation of irradiated fuels and materials is an obstacle. There are few remaining test reactors. In addition, fabrication, irradiation testing, and characterization are all done in different countries. A universally licensed cask to transport samples between test and PIE facilities would be very beneficial.
- To enable the collaboration among international facilities, international regulations must be evaluated and updated to facilitate international collaborative testing. It should not be a special effort to transport every sample.

7.5 Measurements

- Sample preparation is important to advanced examination capability; therefore, standards must be developed for sample preparation.
- International standards for TRU materials (e.g. plutonium) must be developed to be able to compare data.
- Some measurements techniques are useful for multiple parameters and the main PIE hotcells should include such standard techniques (a specialists' team can identify such a standard basic capability matrix (e.g. TEM, EPMA, SIMS).
- For some of the novel measurement techniques, no standards exist. The development of an international standard would be needed if data will be shared among the collaborators.
- Periodic round-robin testing on samples among the collaborators will enhance the confidence on the data.
- The ability to perform characterization and PIE for fuel under severe accident conditions (beyond failure) is a new challenge. The capabilities are quite limited in this area.
- There is a certain volume of work that needs to be done to complete the spectrum for thermodynamic specific data, including fresh fuel. A free access international validated thermodynamic database would be beneficial.
- Trace element detection is very limited (SIMS measurements) and we are not ready to generate validated compositions for spent fuels, etc.

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APPENDIX A

International Workshop Participants

Appendix A
International Workshop Participants

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APPENDIX B

Workshop Presentations

Appendix B
Workshop Presentations

#	Presenter	Org/Country	Title
1	K. Pasamehmetoglu	INL, US	Workshop on Characterization & PIE Needs to Support Science-Based Development of Innovative Fuels
2	U. Basak	IAEA, Austria	Joint IAEA HOTLAB Post Irradiation Examination Facilities Database
3	T. Wiss (J. Somers)	ITU, Germany	TEM Facility Development in Europe
4	B. Lee	KAERI, Republic of South Korea	Irradiation Status and PIE Plan of Metallic Fuel for SFR
5	D. Miotla	DOE-NE, US	United States Perspective on Postirradiation Examination Capabilities
6	V. Bouineau	CEA, France	Fuel Performance Code: Characterization and PIE Needs for Modeling Validation
7	M. Freyss	CEA, France	Atomistic Modeling for Innovative Fuels
8	S. Hayes	INL, US	PIE Needs to Support Fuel Design & Safety Testing
9	M. Pouchon	PSI, Switzerland	Beam-line Techniques
10	R. Kennedy	INL, US	Laser-based Measurement Techniques for Nuclear Materials
11	L. Snead	ORNL, US	X-Ray Microtomography
12	J. Lamontagne	CEA, France	Current and Future Micro-analysis Devices in the LECA-STAR Facility
13	F. Sudreau	CEA, France	Post Irradiation Examinations: LECA-STAR Facility
14	D. Papaoiannou (J. Somers)	ITU, Germany	Application of X-ray Absorption to Density Measurements

Workshop on characterization & PIE Needs to support science-based development of innovative fuels

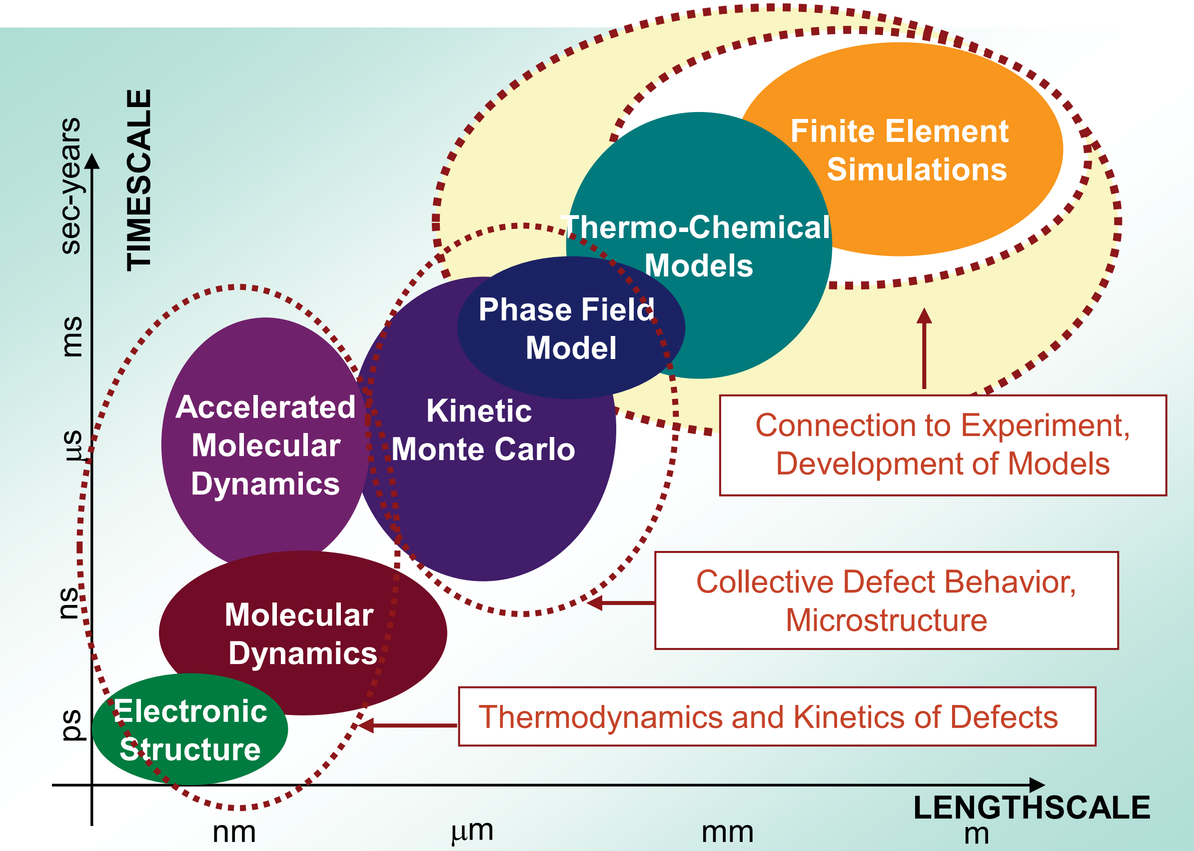
Kemal Pasamehmetoglu

June 16, 2011 (Paris, France)

OECD/NEA Expert Group on Innovative Fuels

Background

- Tradition PIE relies on a limited number of measurements mostly at the engineering-scale
- Traditional PIE provides an integral view of the performance parameters, which is an aggregate of multiple phenomena occurring simultaneously
- The science-based approach aimed at a fundamental understanding of the performance phenomena require measurements at lower length- and time-scales
- Techniques and capabilities aimed at such measurements with highly radioactive samples are being developed at multiple institutions internationally



Workshop Objectives

- To review the existing characterization and PIE capabilities and the novel techniques that are being developed at the international facilities

- To identify the gaps to meet the needs of a 21^{st} century characterization and PIE capabilities

Workshop Scope

- **Presentations by experts from various organizations on the status of existing facilities, capabilities and near-term plans**
  - **sample-sharing possibilities, main features and characteristics of the various facilities and the capability of performing out-of-pile testing on irradiated fuels and material samples**
- **Presentations on characterization and PIE needs for 21st century fuel development**
  - Fuel designer and safety analyst perspective
  - Modeling and simulation and material science perspective
- **New equipment and novel techniques that are being developed at various institutions**

Panel Discussion and Breakout Session

- Invited experts will comment on
  - how the new capabilities, when available, would transform the fuel development paradigm.
  - the major gaps and obstacle to achieving a new, science-based paradigm for development
  - major areas that were not covered in the presentations that should be focused on during the breakout sessions
- 2 breakout sessions
  - Engineering scale and micro-structure characterization (> 1 μm)
  - Lower length scale characterization (< 1μm)

IAEA Activities:

Joint IAEA HOTLAB Post Irradiation Examination Facilities Database

Presented by

U.Basak

Nuclear Fuel Cycle and Materials Section

International Atomic Energy Agency

IAEA

Integrated Nuclear Fuel Cycle Information System

Web based system with public access from 2001

Initially based on the Agency's electronic databases created in 1990s

Information collected from contact points in Member States, consultants and open sources.

NFCIS

UDEPO

PIE

NFCSS

MADB

The **iNFCIS** web site is designed as a "one stop" resource for technical and statistical information about nuclear fuel cycle activities worldwide, as reported to the IAEA. The system includes four databases and one computer simulation system published by the IAEA's Nuclear Fuel Cycle and Materials Section in the Division of Nuclear Fuel Cycle and Waste Technology.

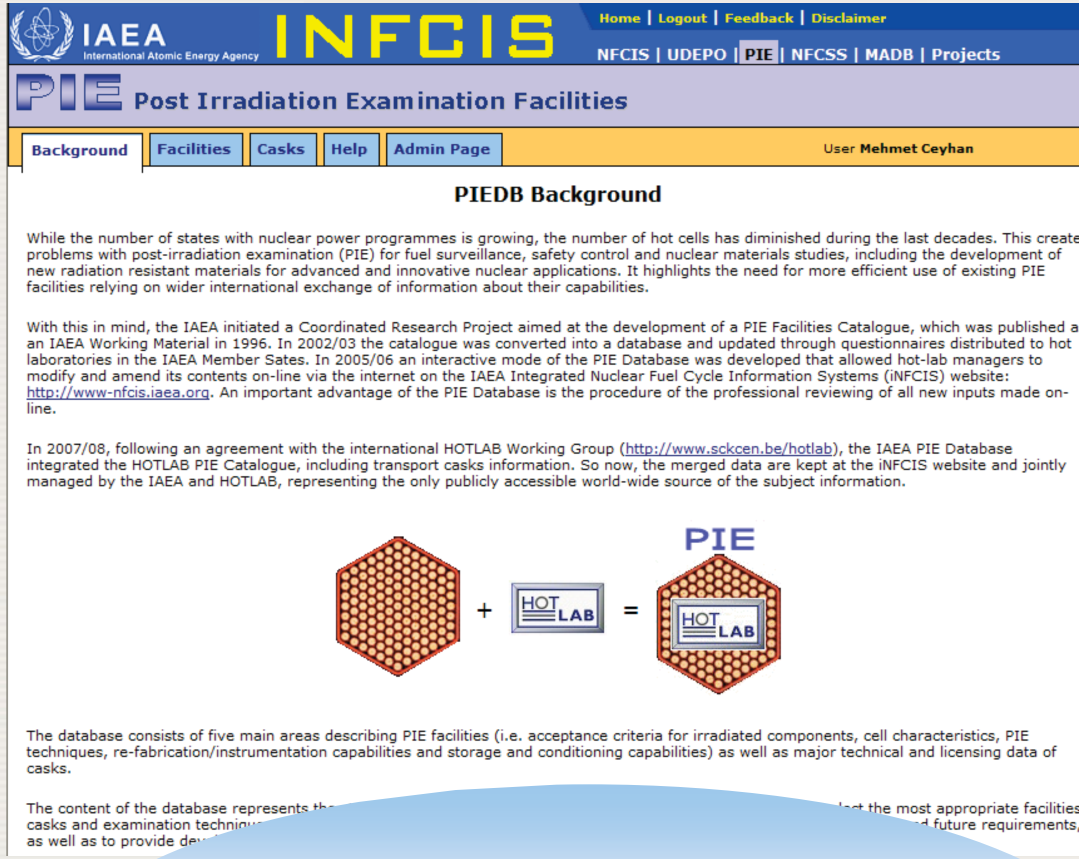
Nuclear Fuel Cycle Information System (NFCIS) covers civilian nuclear fuel cycle facilities around the world. It contains information on operational and non-operational, planned, and cancelled facilities. All stages of nuclear fuel cycle activities are covered, starting from uranium ore production to spent fuel storage facilities.	NFCIS
World Distribution of Uranium Deposits Database (UDEPO) covers uranium deposits around the world, drawing on reports to IAEA technical meetings and other sources. It includes classification of deposits, technical information about the deposits, detailed geological information about regions, districts and deposits.	UDEPO
Post Irradiation Examination Facilities Database (PIE) is derived from a catalogue of such facilities worldwide that the IAEA issued in the 1990s. It includes a complete survey of the main characteristics of hot cells and their PIE capabilities.	PIE HOT LAB
Nuclear Fuel Cycle Simulation System (NFCSS) is a scenario-based simulation system to estimate long-term nuclear fuel cycle material and service requirements as well as material arisings. The code uses simplified approaches to make estimation.	NFCSS
Minor Actinide Property Database is a bibliographic database on physico-chemical properties of selected Minor Actinide compounds and alloys. The materials and properties are selected based on their importance in the advanced nuclear fuel cycle options	MADB

http://www-nfcis.iaea.org

IAEA

Joint IAEA HOTLAB PIE database: history and status

- Created in 1990s, published as a working material in 1996
- Transformed into an electronic database in 2003
- Merged with EC HotLab Database in 2008, casks information was added from EC HotLab database
- Modification and update 2009/10
- Wider collaboration with HOTLAB and NULIFE
- **Data update by facility co-ordinators directly, through a web form**
- **Verification of new data by the DB reviewers**



42 facilities from 22 countries and 19 casks

Joint IAEA-HOTLAB PIE database: the database

IAEA International Atomic Energy Agency INFCIS

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NFCIS | UDEPO | ThDEPO | PIE | NFCSS | MADB | Projects

PIE Post Irradiation Examination Facilities

Background | Facilities | Casks | Help | Admin Page

User **Alan Aqrawi**

List of PIE Facilities

Technique	Technique Topics	Country
Any	Any	All
Name contains:	Go / Reset All Filters	

Total **42** records found in **3** pages. 1 2 3

Country	Facility Name	#-of-DE Techniques	#-of-NDE Techniques
Argentina	CELCA	3	4
Belgium	LHMA - Laboratory for High and Medium Activity - SCK-CEN, Belgium	25	9
Belgium	SCK•CEN - Chemical and Radiochemical Measurements	3	2
Brazil	CTMSP - Hot Cell Pilot Laboratory	2	5
Canada	Chalk River Laboratories, AECL	16	7
Czech Republic	NRI - Nuclear Research Institute Rez plc - Mechanical Testing Department	5	0
France	AMI - Electricité de France Chinon Laboratory	15	7
France	ATALANTE-alpha workshop, lab., analyses, transuraniens, reprocessing studies	6	1
France	CEA - LEFCA - Laboratory for Study & Experimental Manufacturing of Advanced Fuels	6	1
France	CHICADE Hotlab Facility	0	0
France	LECA - Laboratoire d'Examen de Combustibles Actifs	13	5
France	LECI - Laboratoire d'Etudes des Combustibles Irradiés	24	8
France	STAR - Station de Traitement, Assainissement Reconditionnement	0	7
Germany	AREVA NP GmbH NTCRH-G Hot Cells	11	3
Germany	EC, JRC Institute for Transuranium Elements - Hot Laboratory	8	9
Germany	FZJ - Forschungszentrum Juelich Hot Materials Lab (HML)	9	1
Germany	FZJ - Forschungszentrum Juelich Large Hot Cells (GHZ)		
Greece	NCSR "DEMOKRITOS" - RADIOCHEMICAL STUDIES AND QUALITY CONTROL		
Greece	NCSR "DEMOKRITOS"- RADIOISOTOPES AND RADIOPHARMACEU		
Hungary	MTA-KFKI AEKI "Atomic Energy Research Institute"		

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PIE Post Irradiation Examination Facilities

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User **Alan Aqrawi**

List of Casks

Total **19** records found in **1** pages. 1

Cask Type	Cask Provider
AGNES Cask	LA CALHENE
BG 18	TRANSNUBEL
Croft GB2767B	CROFT
Croft GB2835A	CROFT
CTB	CEA
IR 500	CEA
IU 25	CEA
NCS 45	NUCLEAR CARGO + SERVICE GmbH
PADIRAC RD15IIB	LA CALHENE
PADIRAC RD20IIB	LA CALHENE
PN-UO2	CEA
TN TM – MTR	COGEMA LOGISTICS
TN TM – UO2	COGEMA LOGISTICS
TN TM 106	COGEMA LOGISTICS
TN TM GEMINI	COGEMA LOGISTICS
TNB145	TRANSNUBEL
TNB170	TRANSNUBE
TN-BGC 1 version B(M)	
TN-BGC 1 version B(U)	

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IAEA

Joint IAEA-HOTLAB PIE database: webform

1. Register as a contributor with the DB.
2. On-line entry of data by a facility coordinator
3. Verification of new data by the DB reviewers.
4. Release and integration of data into the DB.

IAEA

Joint IAEA-HOTLAB PIE database: content

The DB consists of 5 main areas describing PIE facilities:

- Acceptance criteria for irradiated components.
- Cell characteristics.
- PIE technology.
- Refabrication/instrumentation capabilities.
- Storage and conditioning capabilities.

As well as major technical and licensing data of casks.

Joint IAEA-HOTLAB PIE database: content

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List of PIE Facilities

Technique	Technique Topics	Country
Any	Any	All
Name contains:	Go / Reset All Filters	

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France	ATALANTE-alpha workshop, lab., analyses, transuraniens, reprocessing studies	6	1
France	STAR - Station de Traitement, Assainissement Reconditionnement	0	7
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Germany	FZJ - Forschungszentrum Juelich Hot Materials Lab (HML)	9	1
Germany	FZJ - Forschungszentrum Juelich Large Hot Cells (GHZ)		
Greece	NCSR "DEMOKRITOS" - RADIOCHEMICAL STUDIES AND QUALITY CONTROL L		
Greece	NCSR "DEMOKRITOS"- RADIOISOTOPES AND RADIOPHARMACEUTI		
Hungary	MTA-KFKI AEKI "Atomic Energy Research Institute"		

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Example: Chalk River Lab, AECL, Canada

#DE-Tech.: 16, #NDE-Tech.: 7

IAEA

Joint IAEA-HOTLAB PIE database: content

The DB consists of 5 main areas describing PIE facilities:

- General information & cell characteristics
- Acceptance criteria for irradiated components.
- PIE technology.
- Refabrication/instrumentation capabilities.
- Storage and conditioning capabilities.

As well as major technical and licensing data of casks.

IAEA

Joint IAEA-HOTLAB PIE database: content

Export to PDF

PIE Facility Report

Facility Name : Chalk River Laboratories, AECL

Export to PDF

General & Cell Charac. | Acceptance Info. | Techniques | Refabrication & Instrumentation | Storage & Conditioning | Reference Documents

General

Facility Name	Chalk River Laboratories, AECL	**IAEA Ref #**	3-PIE
Country	Canada	**Last Update**	2010
Address	Chalk River Laboratories, St. 17, Chalk River, Ontario, KOJ-1J0, Canada		
Contact Person	Mr Allen Bakewell (Nuclear Facilities Operation Manager)		
Second Contact Person			
Phone	613-584-8811 ext 46241	**Fax**	
Email	bakewella@aecl.ca		
Web Address	www.aecl.ca	(Please notify us if you can not reach this web address!)	
Additional Information			

Last Update	2010

Cell Characteristics

Purpose	PIE of fuel rods and assemblies		
Gamma Activity Limit (Concrete) (TBq)	37000	**# of Concrete Cells**	11
Gamma Activity Limit (Steel) (TBq)	0		
Gamma Activity Limit (Lead) (TBq)	0		
Cell Atmosphere	Air		
Largest Cell Width (m)	1.8		
Largest Cell Length (m)	4.9		

IAEA

Example: Chalk River Lab, AECL, Canada

General Info & Cell Charact.

Joint IAEA-HOTLAB PIE database: content

The DB consists of 5 main areas describing PIE facilities:

- General Information & Cell characteristics
- Acceptance criteria for irradiated components.
- PIE technology.
- Refabrication/instrumentation capabilities.
- Storage and conditioning capabilities.

As well as major technical and licensing data of casks.

Joint IAEA-HOTLAB PIE database: content

General & Cell Charac. | Acceptance Info. | Techniques | Refabrication & Instrumentation | Storage & Conditioning | Reference Documents

Available Techniques

Technique
Hydrogen Analysis
Visual Examination
Length and Diameter
Gamma Scanning
Oxide Thickness
Rod Puncture
Oxide Thickness
Optical Microscopy
SEM
Image Analysis
Micro-coring
Alpha-Beta Autoradiography
Burnup
Clad Chemical Composition
Tensile Testing
Clad Creep Testing
Tube Burst Testing
Micro Gamma Scanning
TEM
Fission Gas Diffusivity
X-ray Diffraction
O to M Ratio
Specific Heat

Technical Information

Technique	Hydrogen Analysis
Type	DE
Technique Topics	;
Short Description	Measurement by hot vacuum extraction. The (Zy) sample is heate spectrométry analysis at another facility in Saclay.
Form of Data Presentation	report
Comment	Commissioned before 1990, but modified in 2001-2002: new glove
References	T. Bredel, "Determination of hydrogene content in irradiated zircal
Equipment	designed in the laboratory
Standards	yes, Ti 200 ppm,
Test Parameters	1200°C, 30 min (for Zr)
Type of Specimen	Zr clad, Al, stainless steels
Measured Parameters	gas analysis
Calculated Parameters	H2 content in the clad
Features	
Technique Documents	

Example: Chalk River Lab, AECL, Canada

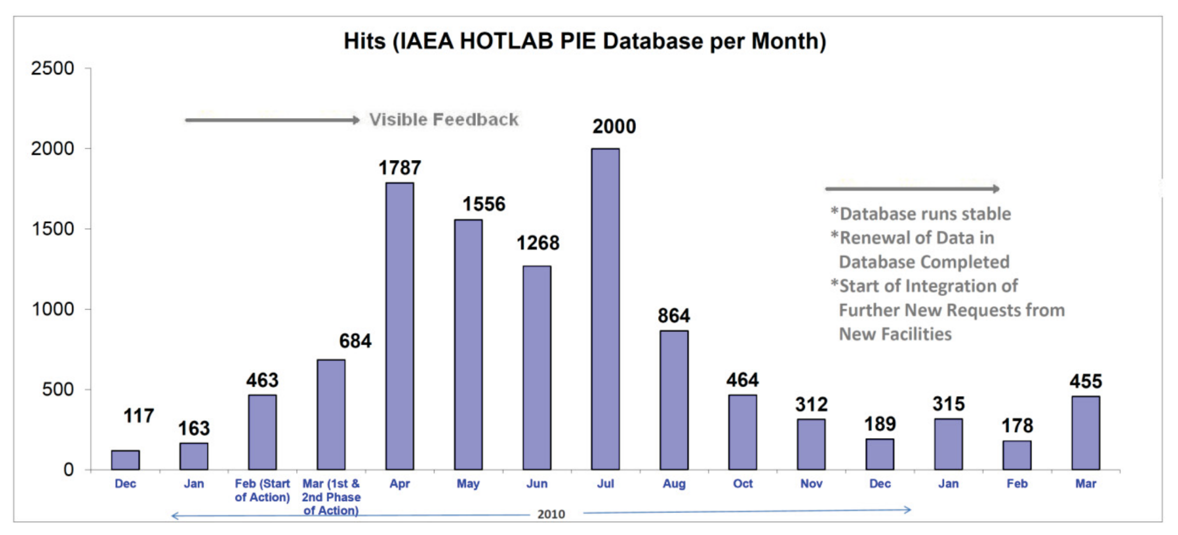
Techniques

IAEA

Joint IAEA-HOTLAB PIE database: growth & statistics

Year-by-year growth in popularity:

- Growing collaboration with HOTLAB (http://hotlab.sckcen.be/)
- In 2010, involvement of the European NULIFE research network for plant life management (launched under the EURATOM framework programme): Instead of developing a new DB, the existing PIEDB is being used and NULIFE facilities are being included.
- Jan-Nov 2010 renewal phase started and completed; with very positive results.



Joint IAEA-HOTLAB PIE database: growth & statistics

Names of contributors to the database:

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Inozemtsev, V. (IAEA, DB Custodian)

Jenssen, H. (External Reviewer)

Leenaers, A. (HOTLAB)

Tulsidas, H. (IAEA, Reviewer)

IAEA

Joint Research Centre (JRC)

TEM Facility Development in Europe

T. Wiss (presented by J. Somers)

ITU - Institute for Transuranium Elements

Karlsruhe - Germany

http://itu.jrc.ec.europa.eu/

http://www.jrc.ec.europa.eu/

Hot Cell facility development in Europe

Refurbishment + replacement of ageing equipment

- EPMA (CEA, JRC-ITU, SCK CEN
- SEM (NRG)
- SIMS (CEA, JRC-ITU)
- TEM (JRC-ITU)

Future needs and perspectives

TEM of MOX (influence of the plutonium)
- **gas behaviour**
- **microstructure**
- **threshold for restructuring**

Behaviour of helium in storage conditions (MOX)

Distribution of metallic FPs and FGs bubbles (tomography)

Future needs and perspectives

New fuels (UC, (U, Pu)C,...)

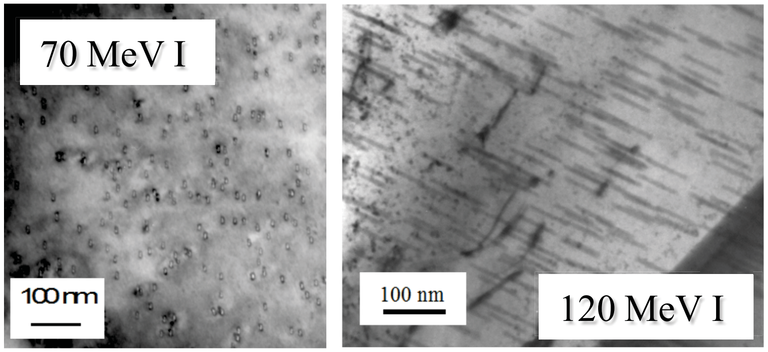
Phase dispersion

Secondary phases (leaching, corrosion, solubilities)

Forensics (microstructural fingerprint)

Nanoparticles

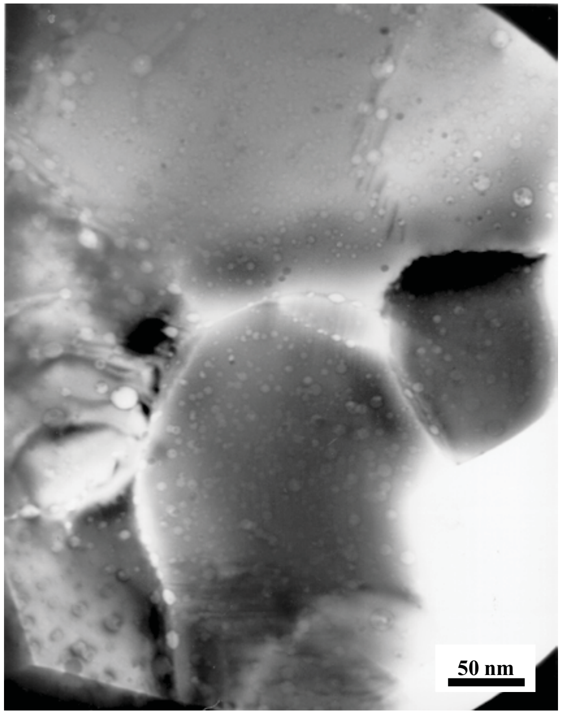
Radiation damage studies



CeO_2

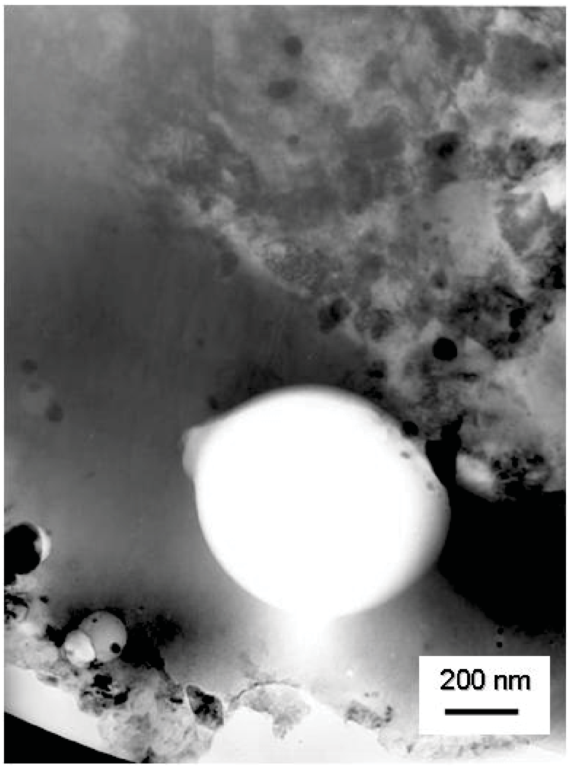
Single effect studies: irradiation with selected ions at given energies

Doping with alpha-emitters for homogeneous damage and helium distribution

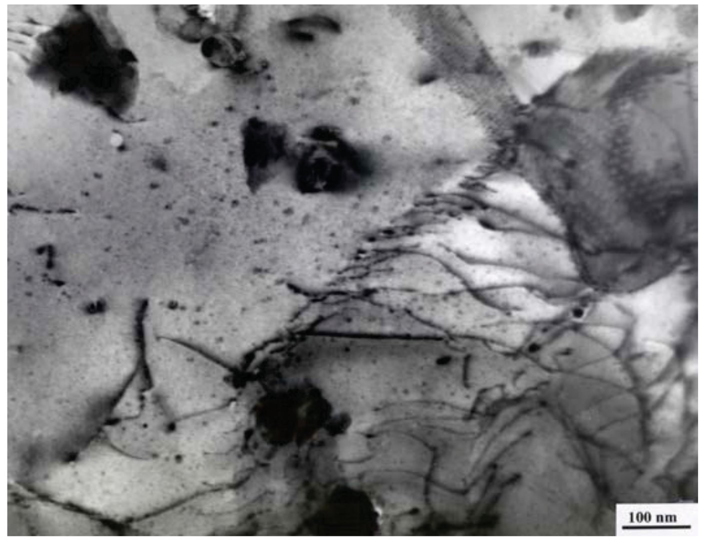


Concomittant effect of different damage sources

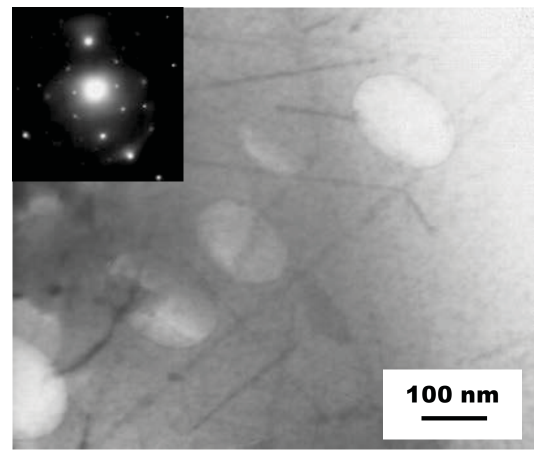
Materials considered



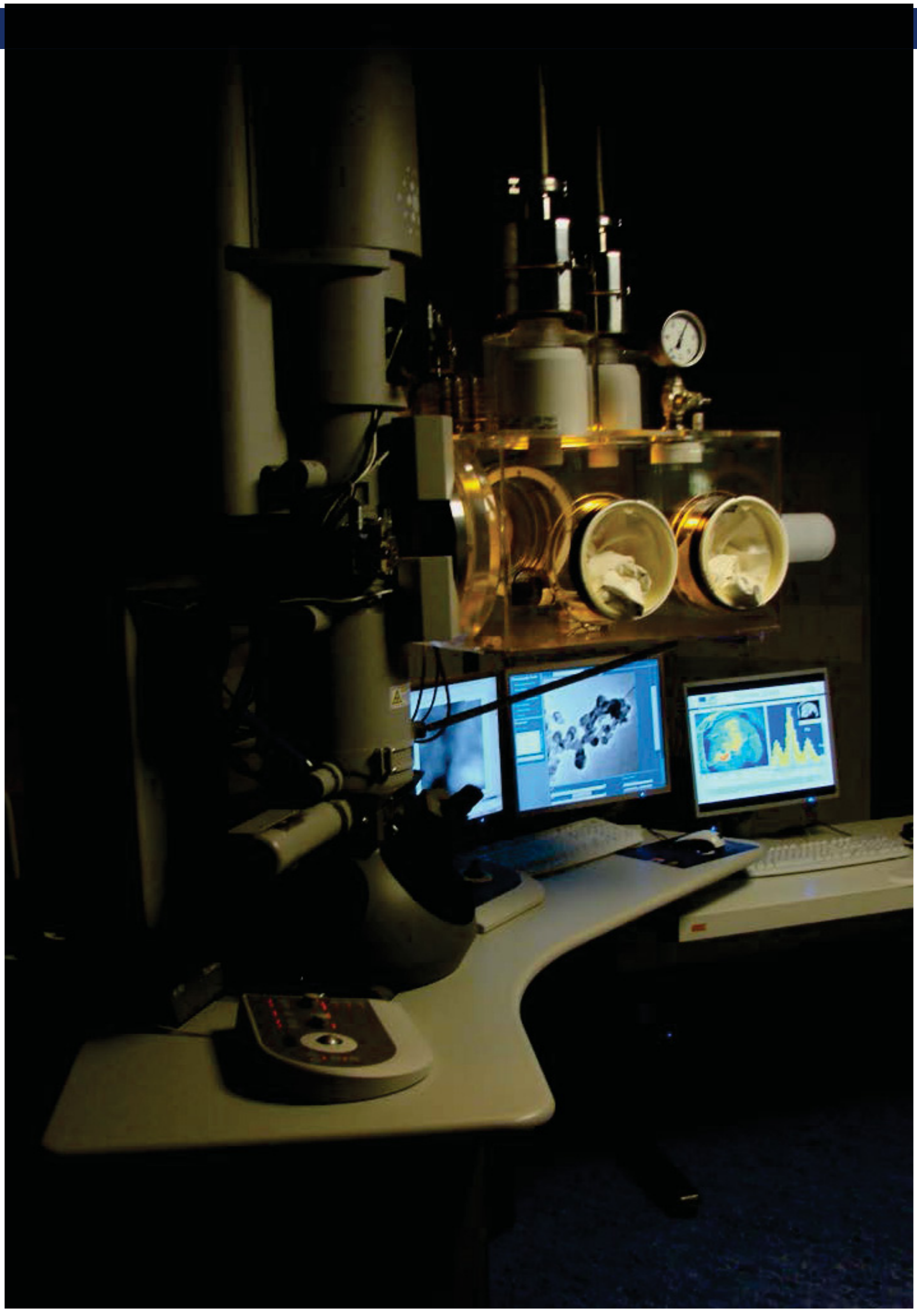
Transmutation targets - IMF



Nuclear fuels



Waste matrices

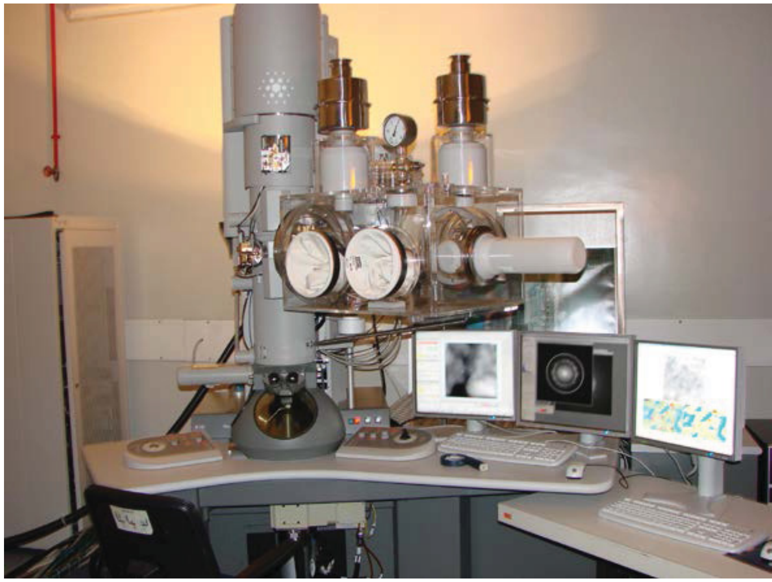


Tecnai G^2 F20 XT

Nearing end of commissioning

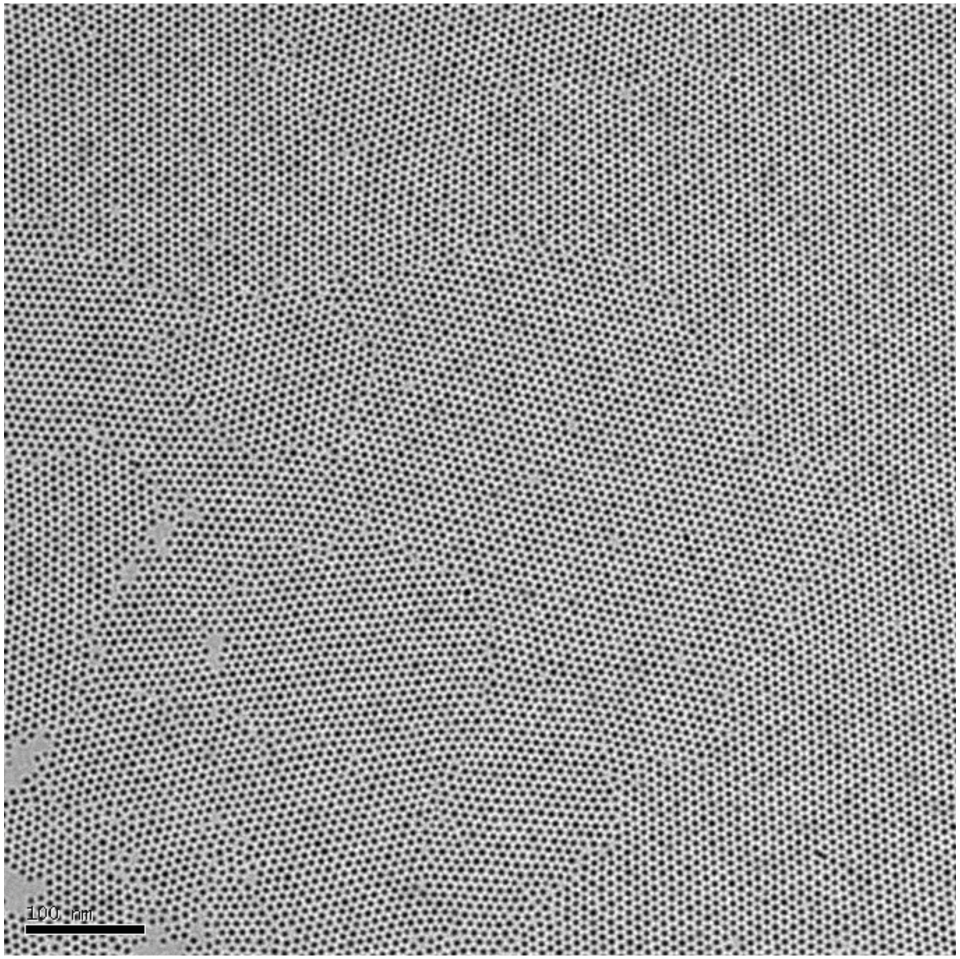
Configuration

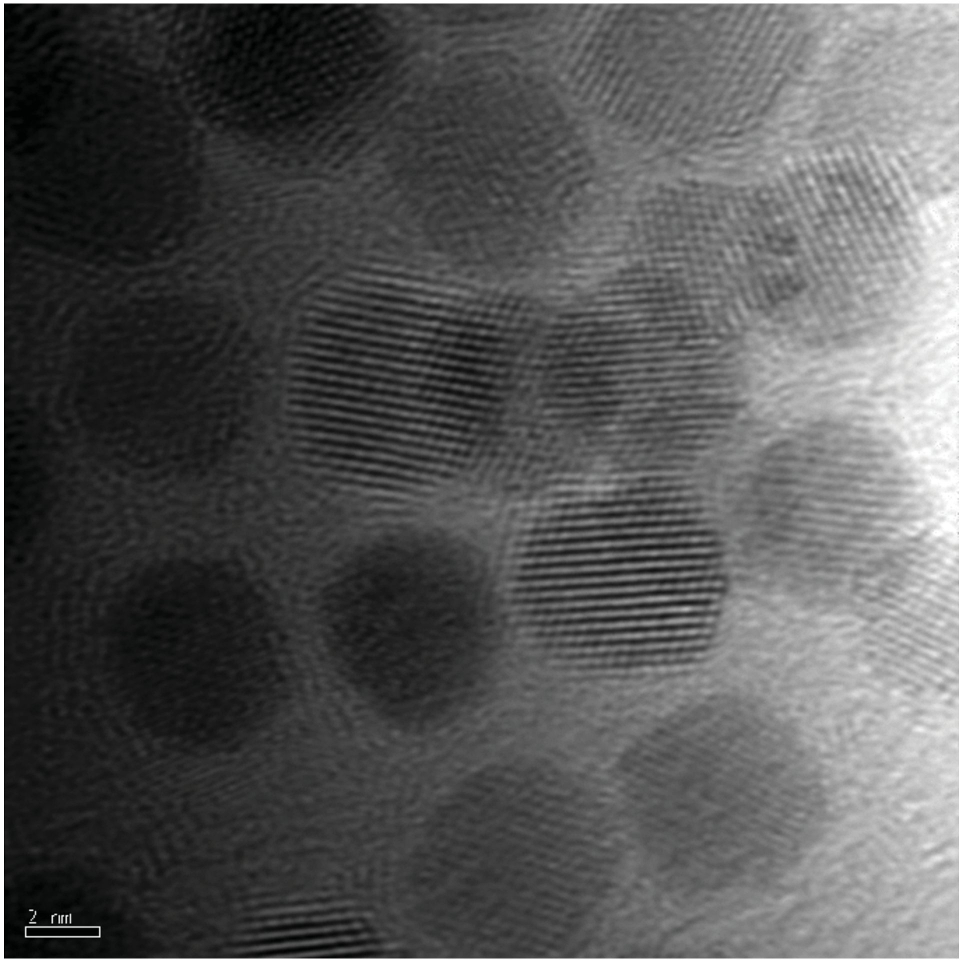
Tecnai™ G² F20 XT



- Field emission gun (high brightness)
- EDX 20 mm^2 , Be-window
- EELS, EFTEM, for light elements
- STEM with HAADF, resolution 0.2 nm
- 3D-tomography (bubbles, crack patterns, phase dispersion, etc)
- 2k-SSCCD camera
- **Remote controlled**
- **Adapted for nuclear materials**

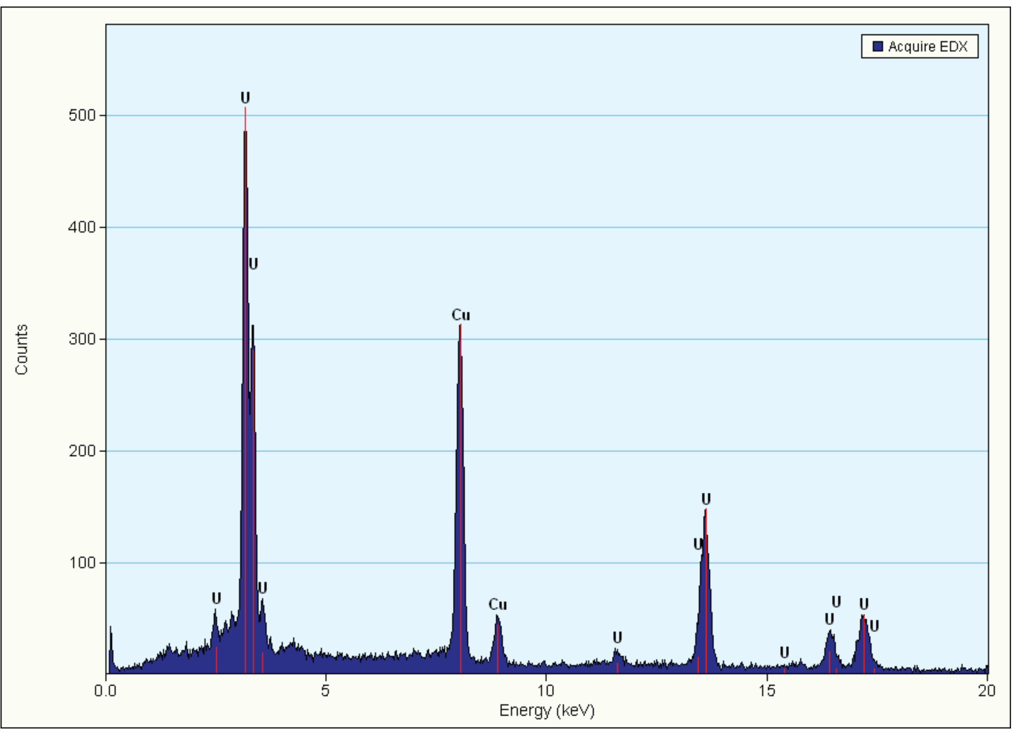
Nanocrystals





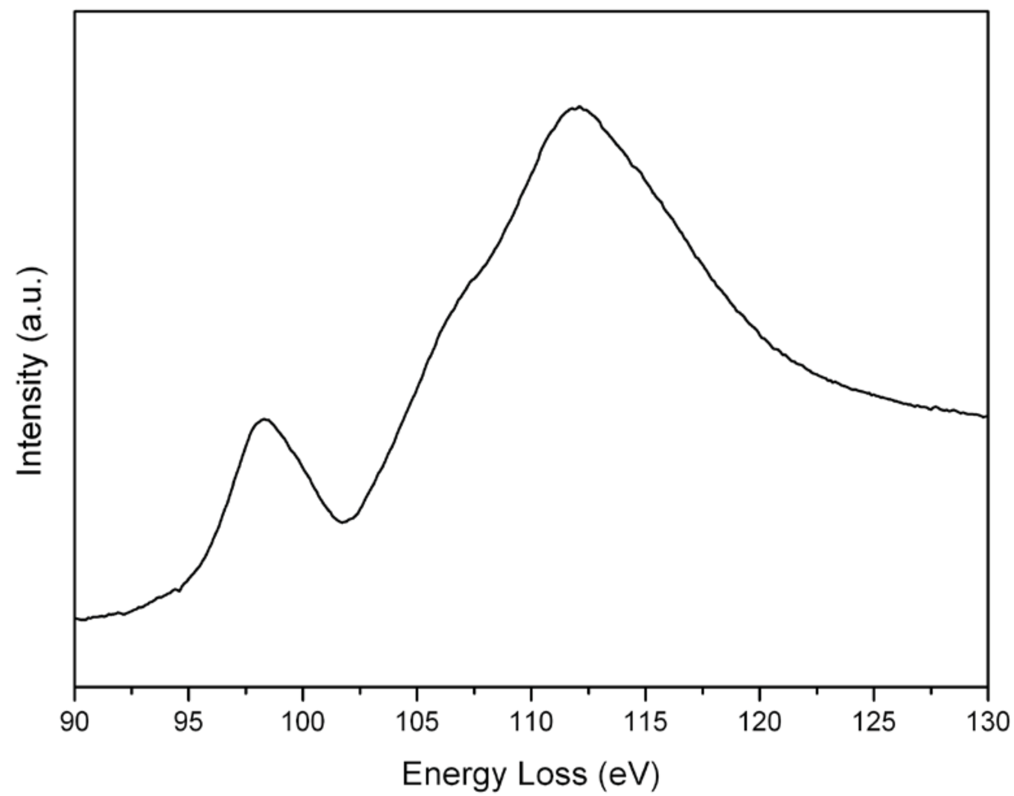
Imaging + elemental analysis and speciation

EDX

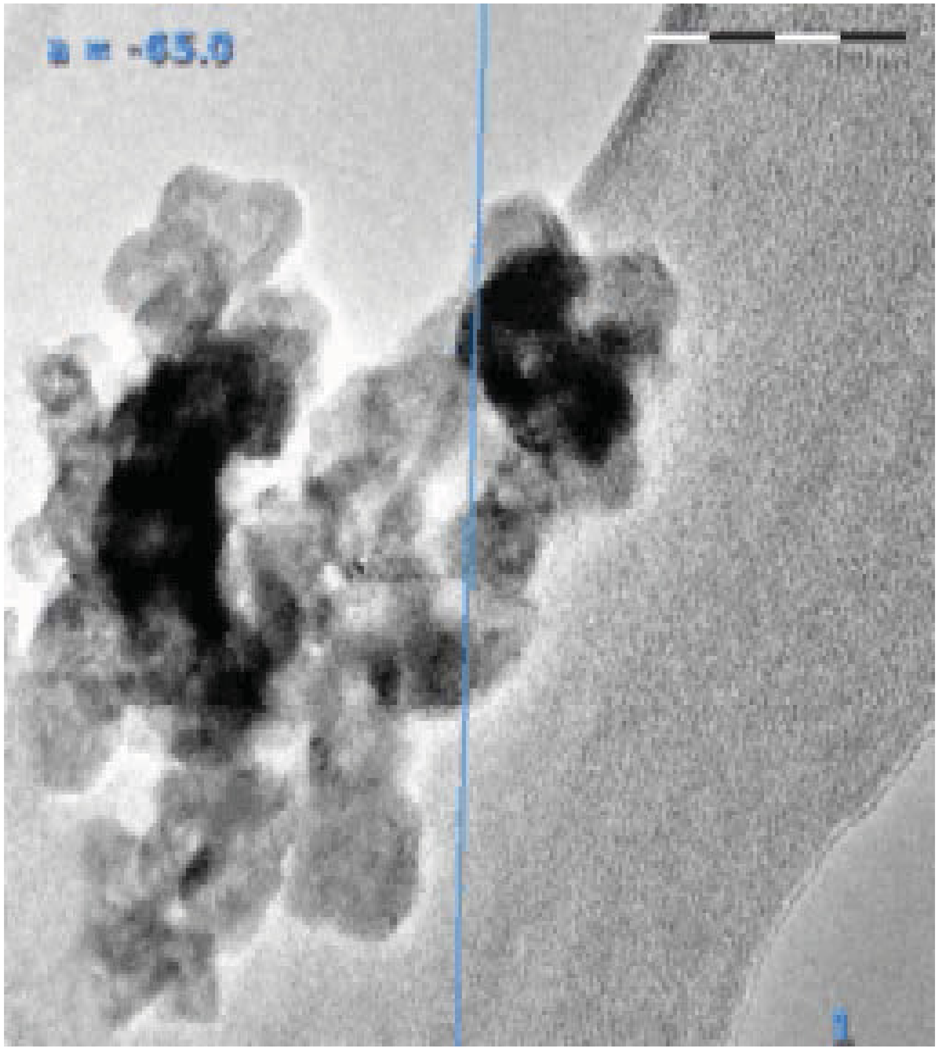


EELS

U $O_{4,5}$ edge

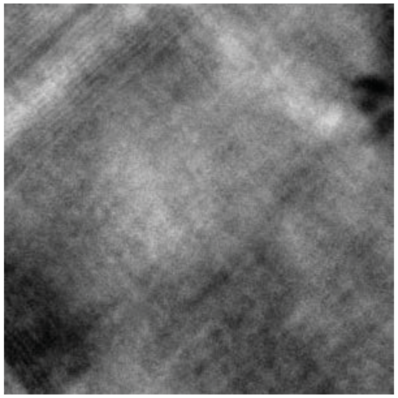


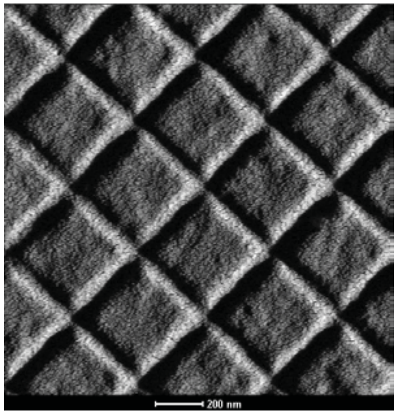
3D Tomography

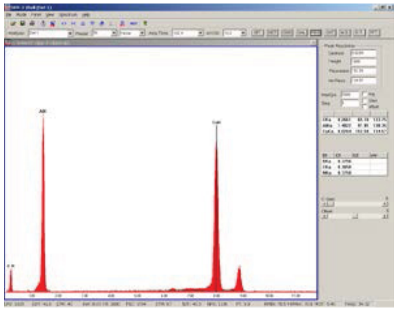


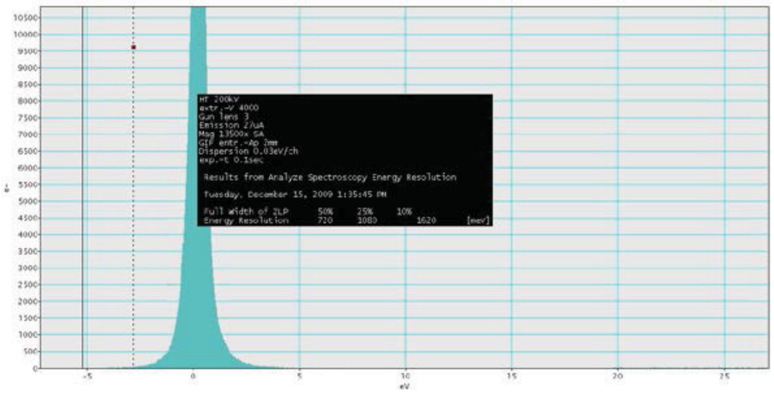
Performance

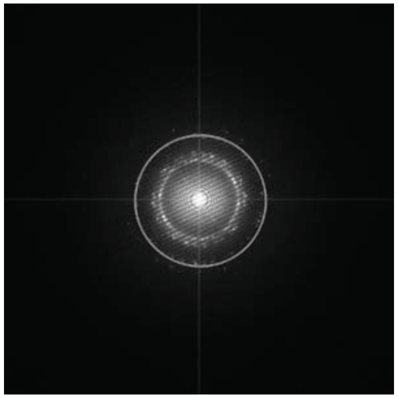
TEM point resolution (nm) 0.25
TEM line resolution (nm) 0.102
Information limit (nm) 0.14
Extended resolution (TrueImage) 0.14
TEM magnification range 22 x - 930
STEM HAADF resolution (nm) 0.23 (0.18)
STEM magnification range 150 x - 230 Mx
Maximum tilt angle -tomography ±70°
EDS energy resolution 134.6 eV
Spot drift 1 $nm.min^{-1}$
Resolution EELS (ZLP) 0.6 eV

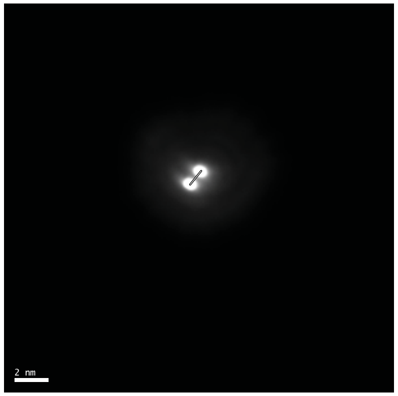












Irradiation Status and PIE Plan of Metallic Fuel for SFR

OECD/NEA Workshop, NEA HQ, France
(characterization and PIE needs to support science-based development of innovative fuels)

2011. 6. 16-17

ByoungOon LEE

KAERI 한국원자력연구원
Korea Atomic Energy Research Institute

OUTLINE

1 INTRODUCTION

2 IRRADIATION STATUS

3 PIE PLAN

4 CONCLUSIONS

Acknowledgments

- **PIE & Radwaste Division**
  - Sangbok Ahn
  - Gilsoo Kim
  - Byungok Yoo
  - Seungjai Baek
  - Woongsub Song
  - Sangyul Baek
  - Woo-Seog Ryu

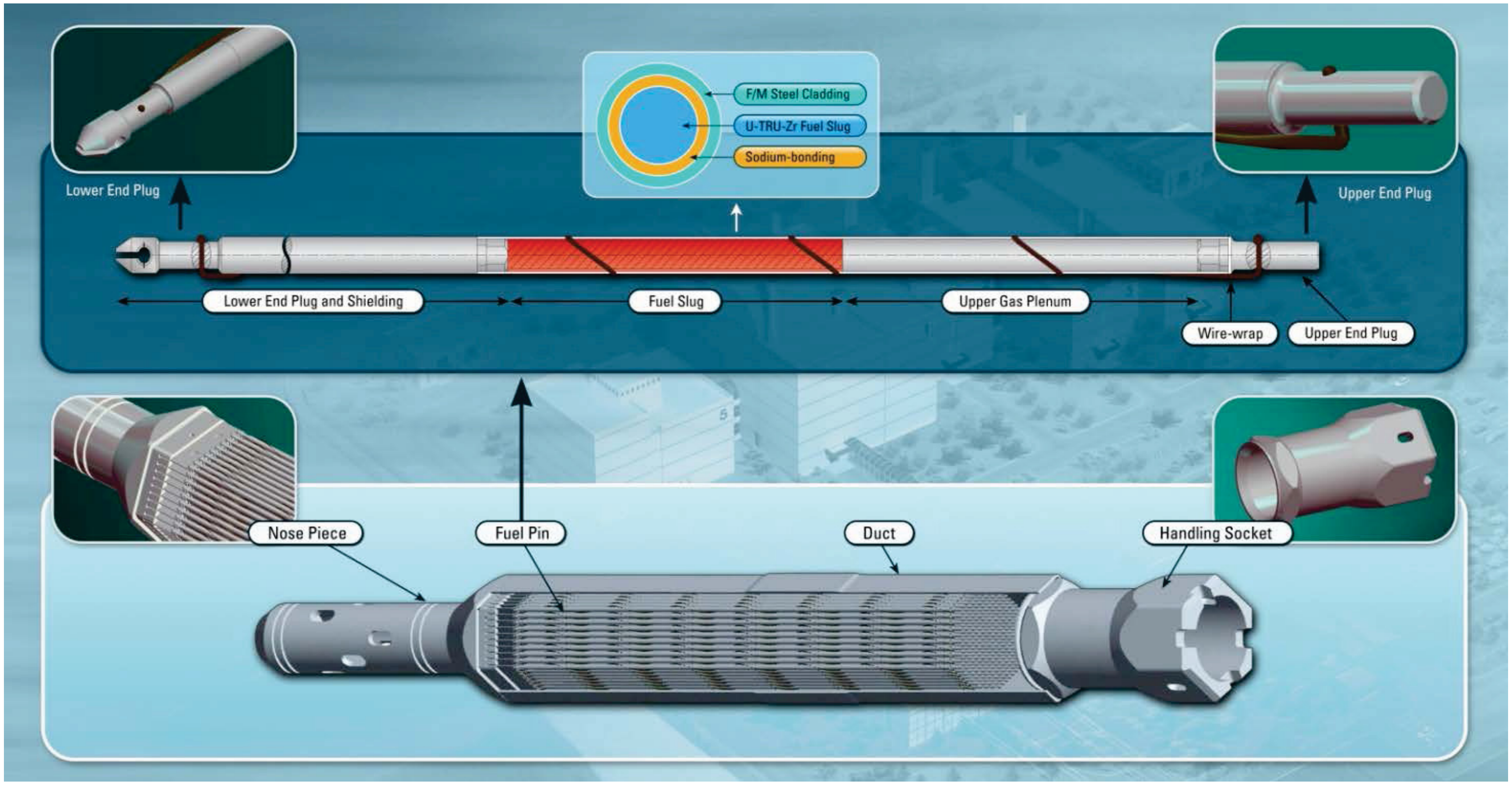
1

INTRODUCTION

Metallic Fuel for SFR

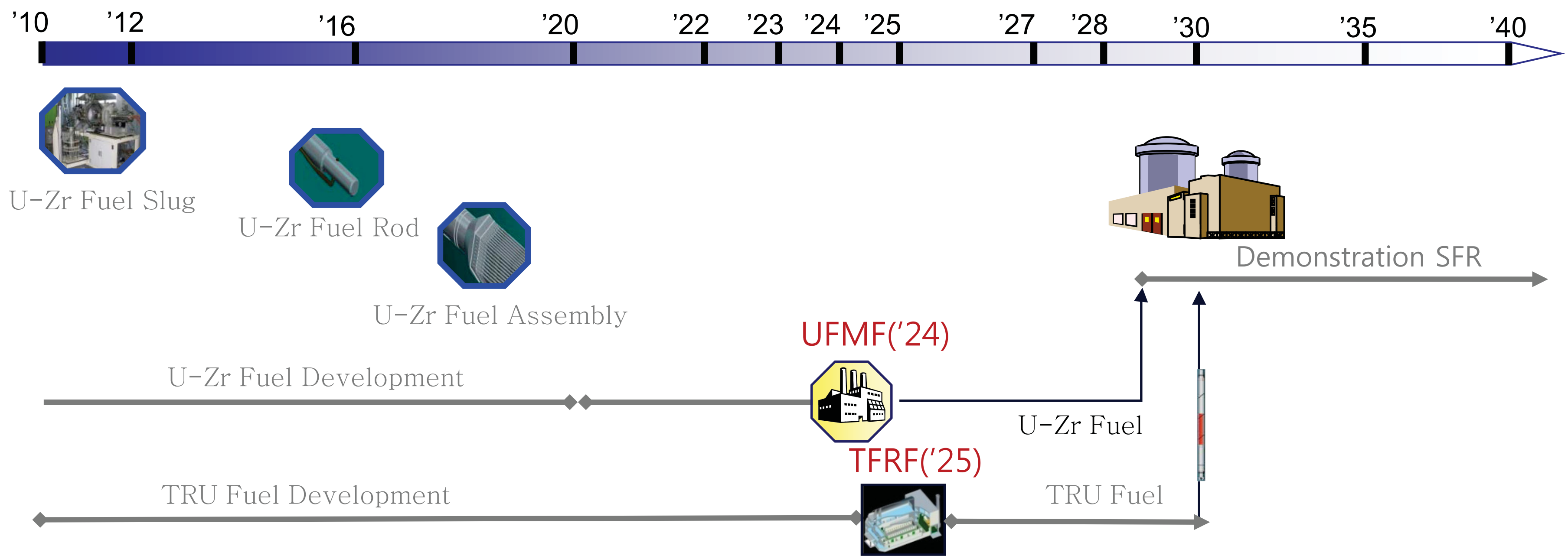
❖ Metallic Fuel Design Concept

- Fuel slug : U-TRU-Zr, U-Zr
- Fuel rod gap filled with sodium
- Cladding and duct : ferritic martensitic steel (HT9)



Metal Fuel Development Plan

- ’07 ~ ’18 : Fabrication technology development of U-Zr fuel
- ‘21 ~ ’28 : Fuel facility construction & fabrication for the SFR plant
- ’30 ~ : U-Zr fuel transition to U-TRU-Zr after performance verification



UFMF: U-Zr Fuel Manufacturing Facility
TFRF: TRU Fuel manufacturing Research Facility

Results of SFR Metal Fuel Development ('07-)

Fuel Fabrication

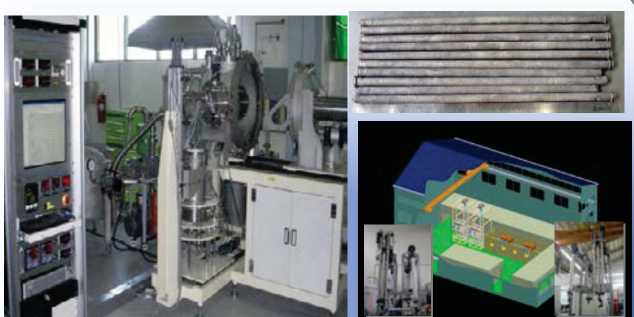
- Design and installation of fuel slug casting system to control volatile elements
- Fabrication of fuel slugs (U-Zr, U-Ce-Zr, U-Mn-Zr)
- Fuel rod fabrication with sodium bonding

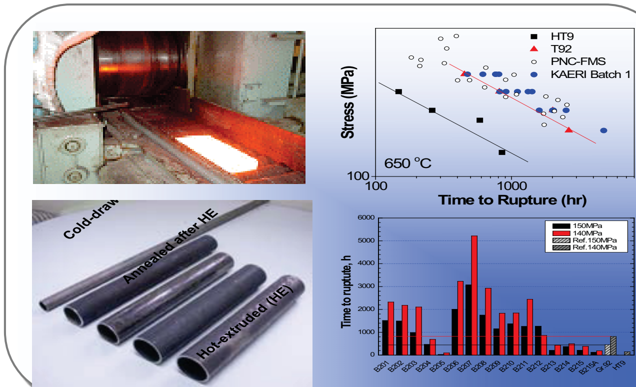
Advanced Cladding

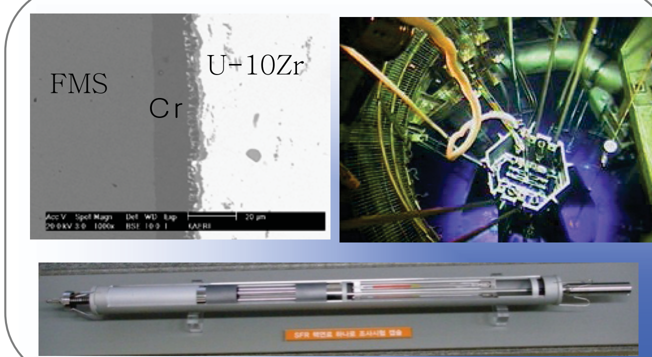
- Design of advanced F/M steel cladding to improve mechanical strengths at high temp. (650°C)
- Better creep strength than conventional HT9 were shown.
- Trial fabrication of conventional HT9 cladding

Performance Evaluation

- Fuel performance analysis and modeling
- Fuel irradiation test in HANARO
- Practical barrier (Cr, V) technology development to prevent fuel cladding chemical interaction



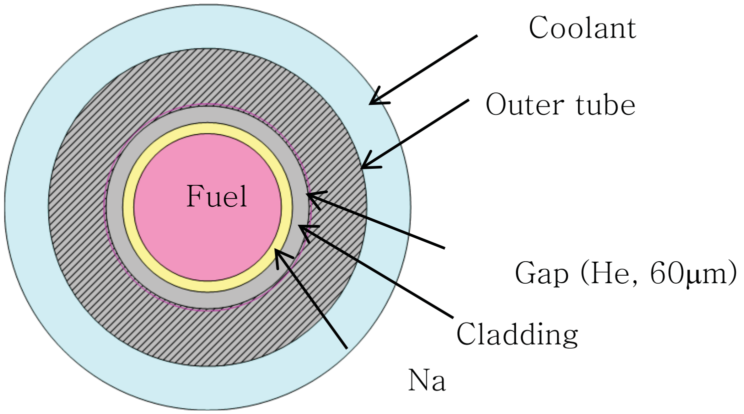




2 IRRADIATION STATUS

Irradiation in HANARO

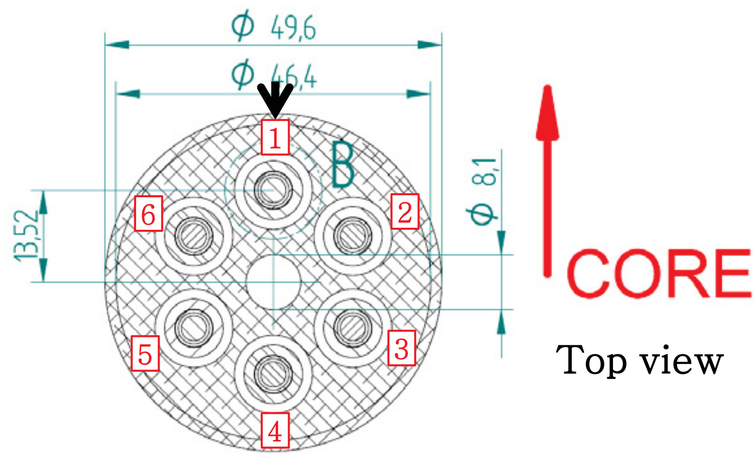
- **2010. 11 : irradiation in HANARO**
- **Irradiation condition**
  - **Fuel : U-10wt.%Zr-(Ce)**
  - **Cladding : FMS**
  - **Fast reactor condition**
    - **Thermal neutron flux filter : Hf**
    - **Fuel temp. control : He gap**
    - **Fuel/cladding gap bonding : Na**

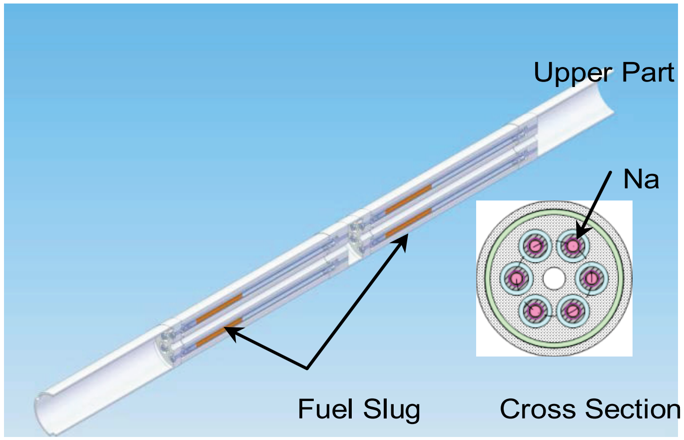


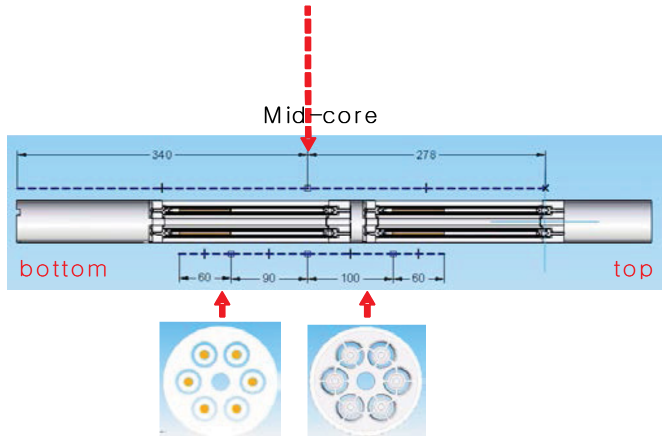
Fuel slug			Cladding			Sealing tube		
OD, mm	Density, g/cc	Length, mm	OD, mm	ID, mm	Length, mm	OD, mm	ID, mm	Length, mm
3.7	15.8	50	5.5	4.6	193	8.62	5.62	233

HANARO Irradiation Test Goal

- Objectives
  - Investigate the effect of design & fabrication variables
  - Examine the level of impurities
    - RE : Ce, Nd, La, Pr
    - Other impurities : C, Si
  - Identify the characteristics of barriers for preventing eutec reaction
    - Barrier material (Cr, V, Zr & Nitride) and performance
- Requirements for 1st irradiation test
  - Ensure the integrity of test fuels and capsule
  - Identify the compatibility of capsule with HANARO core
  - EOL burnup: 3 at%







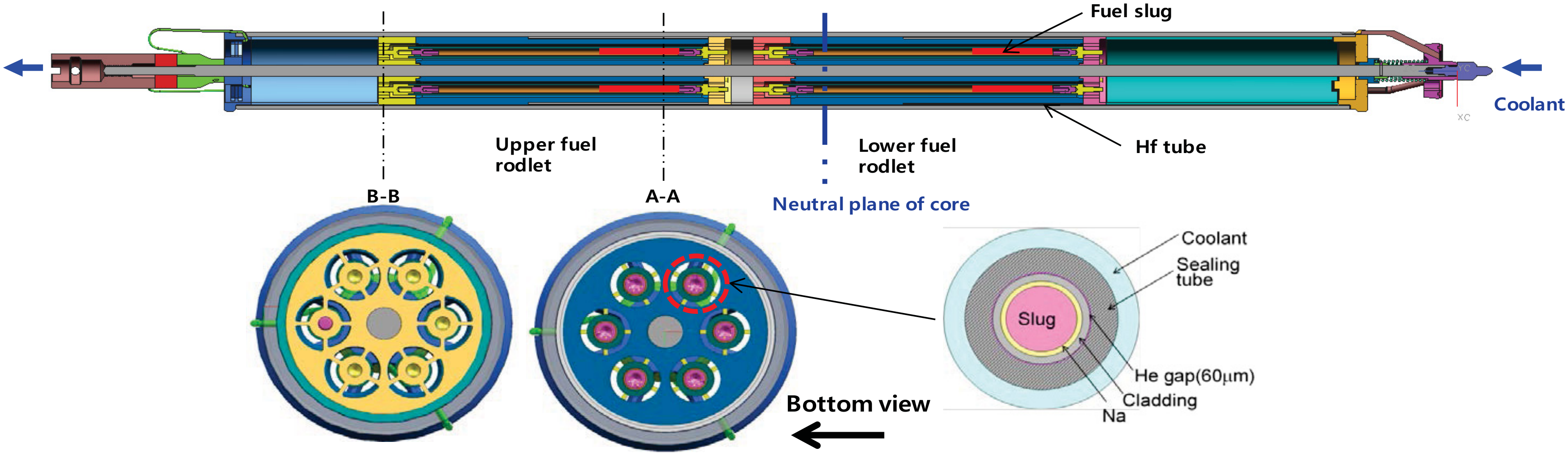
Irradiation capsule model

Irradiation Capsule for HANARO

❑ Characteristics of irradiation capsule

- 12 rodlets : 6 U-10Zr & 6 U-10Zr-5Ce
- Barrier : 20 μm thick Cr
- Cladding : ferritic-martensitic steel (T92)
- Sealing tube : 316L
- Capsule : total length 961 mm, diameter 56 mm, thickness 3 mm
- Hf tubes : upper(1.35 mm) & lower(1.58 mm) to limit fuel linear power

	No.	Fuel slug	Cladding	Barrier
Upper	1	U-10Zr	T92	
	2	U-10Zr	T92	
	3	U-10Zr	T92	Cr
	4	U-10Zr-5Ce	T92	
	5	U-10Zr-5Ce	T92	
	6	U-10Zr-5Ce	T92	Cr
Lower	1	U-10Zr	T92	
	2	U-10Zr	T92	
	3	U-10Zr	T92	Cr
	4	U-10Zr-5Ce	T92	
	5	U-10Zr-5Ce	T92	
	6	U-10Zr-5Ce	T92	Cr

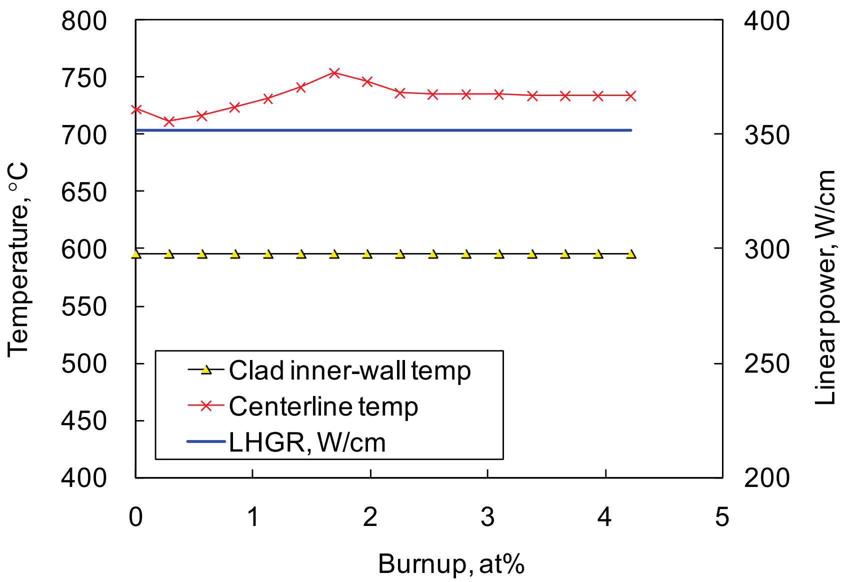


Design of Irradiation Capsule

❖ Preliminary evaluation of fuel performance

– MACSIS code was employed
  - Cladding inner temperature : 600 °C
  - Conservative LHGR : 352 W/cm

– Fuel integrity is maintained during irradiation : temperature, strain



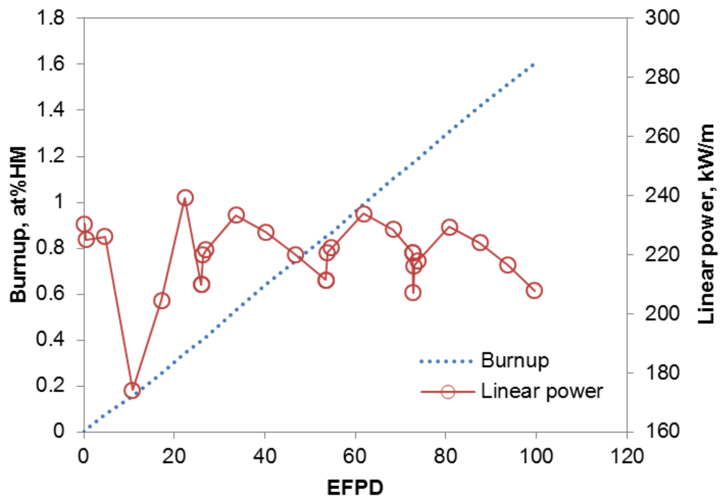
Variation of fuel temperature with burnup

Fuel temperature	– Centerline temperature: 755°C<1235°C – Surface temperature: 600°C<700°C
Cladding Strain, Stress, CDF	– Rod pressure : 12 bar – Stress : 6.7 MPa < 150 MPa – Strain : 0.015 % < 1 % – CDF : 3×10^{-10} < 0.2

Fuel performance analysis with MACSIS code

Status Summary and Planned Activities

- **Status**
  - **Manufacturing of capsule for HANARO irradiation**
  - **Irradiation began ('10.11.15)**
    - **Burnup at 1st irradiation cycle (29EFPD) : 0.3 at.%**
    - **Burnup (2011.3.25, 75 EFPD) : 1.2 at.%**
    - **Burnup (103 EFPD) : 1.6 at%**
- **Planned Activities**
  - **2011. 9~10 : irradiation complete**
  - **PIE ('11~) will begin**
  - **2nd and 3rd U-Zr HANARO irradiation**
  - **U-TRU-Zr ATR irradiation through Joint Fuel Cycle Studies Collaboration**

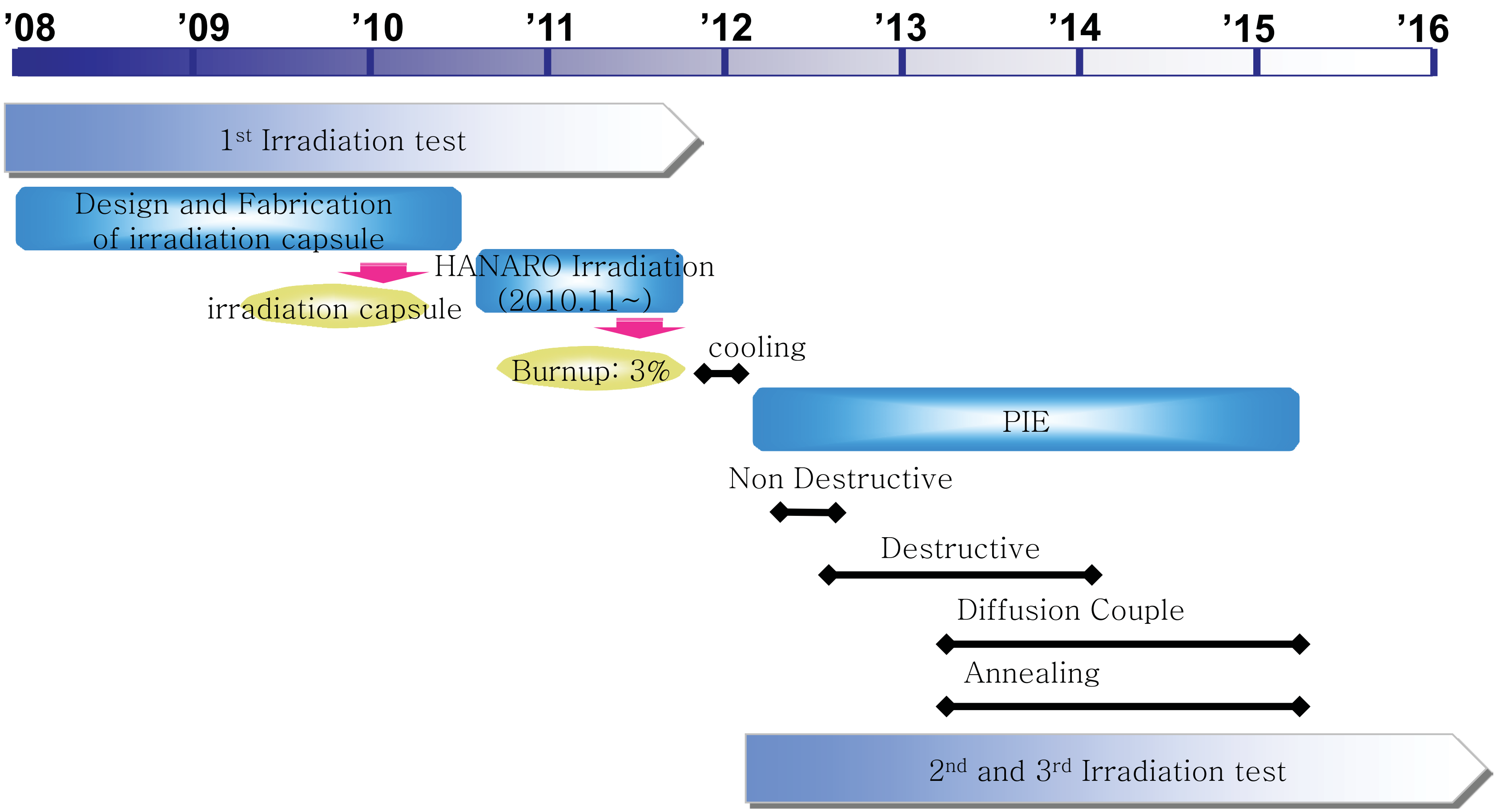


3

PIE PLAN

Irradiation and PIE plan

- **Status & Plan**
  - **PIE will be performed in IMEF hot cell, and techniques and devices are being developed**
    - **Techniques for disassembling rodlets and dealing with sodium in an air cell**
    - **Laser puncturing system for FGR measurement**

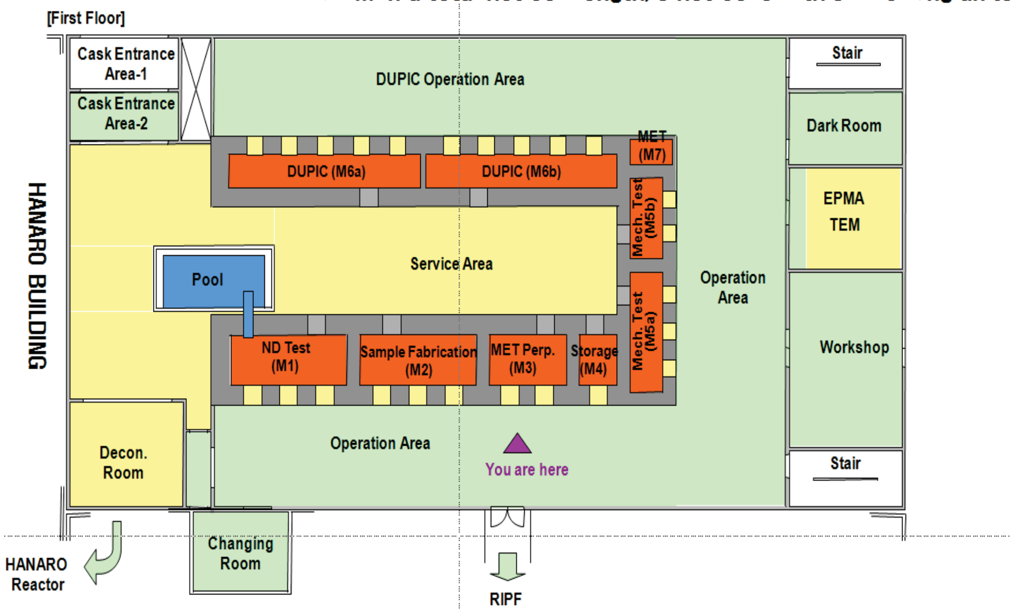


Post-irradiation examination

	Items	Purposes
Nondestructive test	Appearance	• Integrity of fuel rodlet
	Dimension	• Measurement of length and diameter of rodlet
	Gamma scanning	• Variation of burnup along axial direction of rodlet • Axial movement of fission products, especially Cs
Destructive test	Burnup and chemical analysis	• Measurement of fuel burnup with chemical analysis
	Fission gas release	• Variation of FGF with fuel burnup
	Swelling and axial length	• Measurement of variations in swelling and axial length with fuel burnup
	Density	• Measurement of density variation
	Microstructure	• Size and distribution of porosity, distribution of phase and fission product, Infiltration of sodium into fuel
	Constituent redistribution	• Observation of radial constituent redistribution with fuel burnup
	Cladding thickness	• Measurement of cladding deformation with fuel burnup • Measurement of wastage on the inner surface of cladding
	FCCI test	• Fuel-cladding chemical interaction test with irradiated fuel
	Annealing test	• Behaviors of swelling and fission gas release during simulated transient tests with irradiated fuel

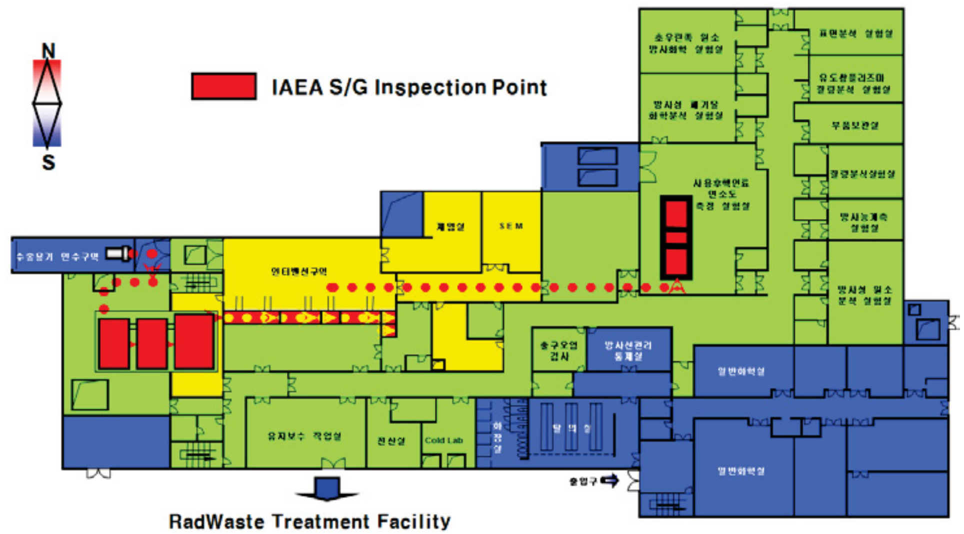
IMEF and PIEF Plan View

- Construction: 1988 yr ~ 1993 yr (26.5M US $)
- 3 stories and a basement (4,000 m^2)
- 71 m in a total hot cell length, 8 hot cells with 31 working units



1st floor plan view in IMEF

3 pool, 4 concrete cell, 2 lead cells

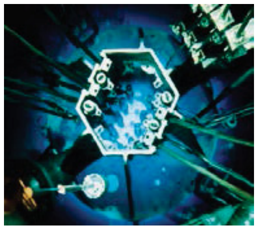


1st floor plan view in PIEF

The main mission of IMEF is to provide PIE services for the irradiated fuels and materials in the HANARO.

And, PIEF is essentially employed for testing and then evaluating the performance and the integrity of nuclear fuels discharged from reactors.

IMEF and PIEF PIE Flow

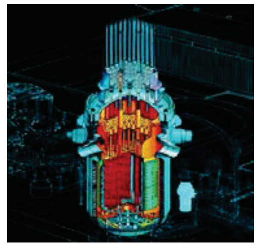


Fuel Capsules
Operated Fuels

HANARO
POWER REACTOR

Irradiated Fuels
Operated Reactor Structure Parts

Transportation/
Unloading



Material Capsule
Opearated Core Parts

<Nondestructive Exam>
- Visual Inspection
- Dim. Measurement
- X-Ray radiography
- Gamma Scanning
- Blistering Test

<Dismantle & Fabrication>
- Capsule Dismantling
- Assembly Dismantling
- Rod Cutting

<Metallographic Sample>
- Resin Impregnation
- Grinding/Polishing
- Etching

<Physical & Mech. Test>
- Density
- Thermal Diffusivity
- Fission Gas Collection
- Hardness

<Meatall. & Micro Analysis>
- Metallographic Investigation
- EPMA
- SEM
- TEM

<Nondestructive Exam.>
- Visual Inspection
- Dimension measurement
- Eddy Current
- irradiation Growth Meas.

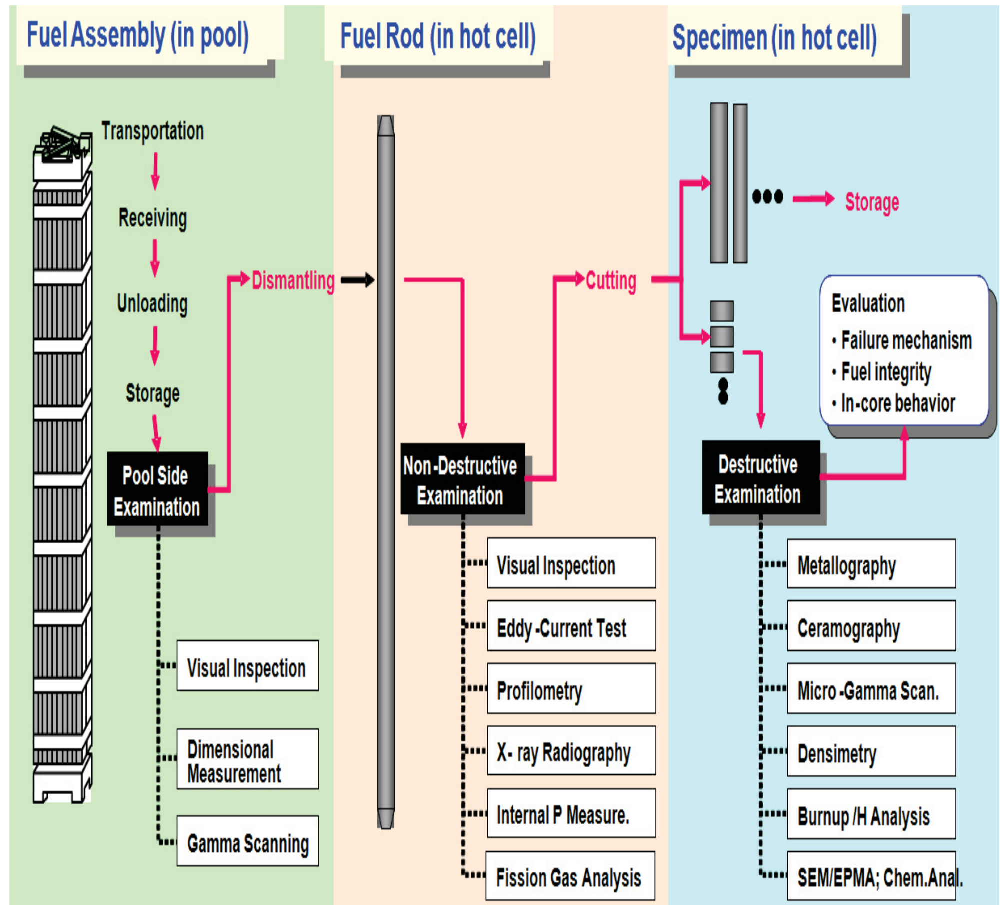
<Dismantle & Fabrication>
- Capsule Dismantling
- Cutting
- Milling
- Electric Discharging

<Mechanical Test>
- Tensile
- Impact
- Fracture & Fatigue
- Compression
- Hardness test

<Metallographic Sample>
- Resin Impregnation
- Grinding/Polishing/Etching

<Physical Test>
- Thermal Diffusivity

PIE flows for HANARO Capsules &
NPP core parts
in IMEF



PIE flows for the operated PWR
fuels from NPP
in PIEF

Hot cell specifications and equipments in IMEF

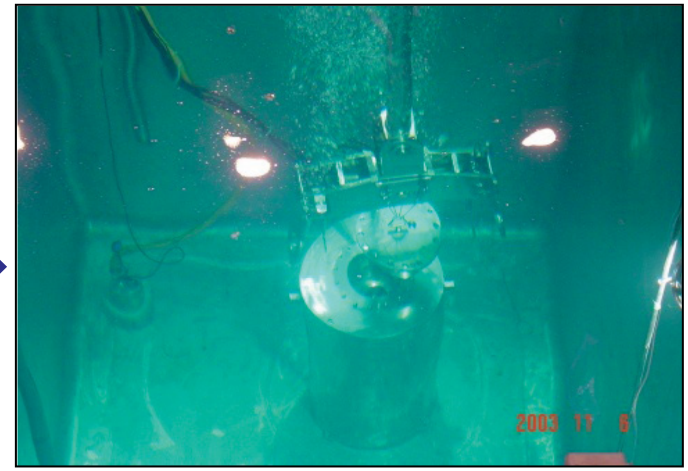
Pool/ Cell	Function	Inside Dimension (m)	Wall Thickness	Windows (ea)	Major Equipment
Pool	Receiving Cask	6.0 x 3.0 x 10.0 (depth)	-	-	-Bucket Elevator
M1 Cell	NDT	7.0 x 3.0 x 6.0	Heavy Conc. 1.2 m	3	- Eddy Current - Gamma Scanning System - X-ray Radiography System - Dia. Measurement System - Rod Puncturing & Fission Gas Collection System
M2 Cell	Specimen fabrication	7.0 x 3.0 x 6.0	"	3	- CNC Machine - Capsule Cutting Machine - Cookie Electric Discharge Machine (EDM)
M3 Cell	Metallographic sample prep.	4.7 x 3.0 x 6.0	"	2	- Low speed saw - Mounting Press - Grinder/Polisher - Periscope
M4 Cell	Sample storage	2.3 x 3.0 x 6.0	"	1	- Specimen Storage Rack
M5a Cell	Mechanical Test	7.1 x 2.0 x 4.0	Heavy Conc. 0.8 m	3	- Impact Tester - Tensile tester for small specimen - Highscope Dimension Machine
M5b Cell	Mechanical Test	4.8 x 2.0 x 4.0	"	2	- Dynamic Tensile Tester - Static Tensile Tester
M7 Cell (M7a, b)	Metallographic Observation	1.5 x2.6 x 4.65	Lead 0.2 m	2	- Optical Microscope - Micro Hardness Tester - Micro balance - Mini SEM
Hot Lab-I	Micro Analysis	5.8 x 7.5 x 8.4	Normal Conc.	-	- EPMA, TEM
Hot Lab-II	Small Specimen Testing	6.0 x 7.0 x4.55	Normal Conc.	-	- Wire EDM - Thermal Diffusivity Tester - Glove Box - Internal Burst Tester - Creep Tester
M6 Cell (M6a, b)	Sequential Progress Test	23.4 X 2.0 X 4.0	Heavy Concrete 1.1 m	10	- Use by DUPIC Project - OREOX Furnace, Mill, Roll Compactor, Mixer, Sintering Furnace, Center less Grinder, Off Gas Treatment
M8 Cell (M8a, b)	Sequential Progress Test	10.3 x 2.0 x 4.3	Heavy Concrete 0.9 m	5	- Use by ACP Project - Slitting Machine, Vol-oxidizer, Metallizer, Smelter, Waste MS Treatment Device, Off Gas Treatment System

Procedure and Functions in IMEF

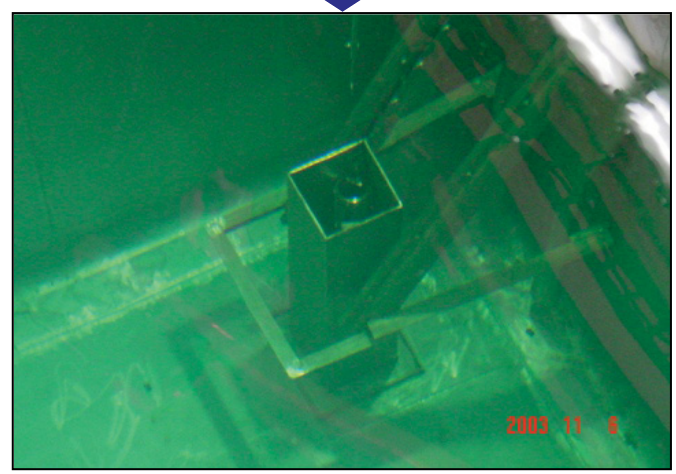
❑ Pool

Cask on moving cart

Moving cask by crane

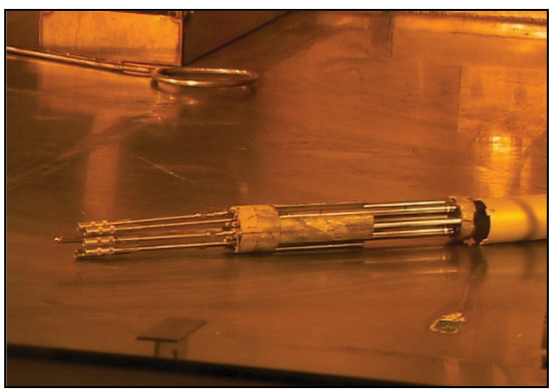


Moving cask into pool



Capsule in the bucket

Hotcell (M1)

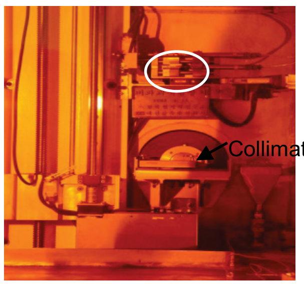


Dismantling capsule

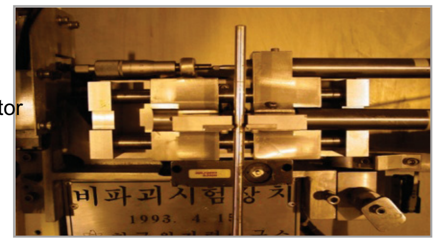
Functions in IMEF

❑ M1 hot cell

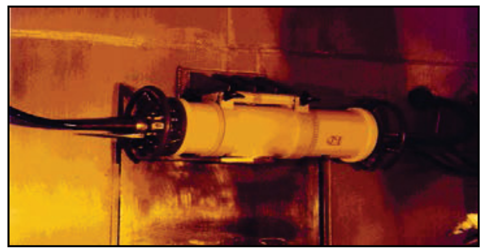
- Main Function: Loading into the Irradiated Capsule, Nondestructive Examination for Fuel Rods
- Equipments: Multipurpose Test Bench (Gamma Scanning, LVDT, Eddy Current Coil)
  X-Ray Radiography System, Annealing Furnace Etc



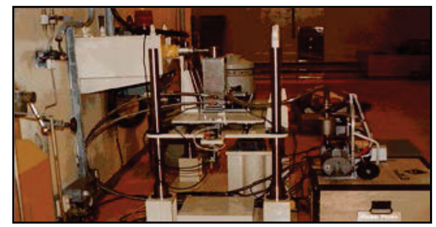
Gamma Scanning System



Diameter Measurement System



X-ray Radiography System

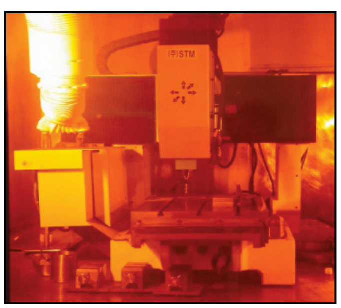


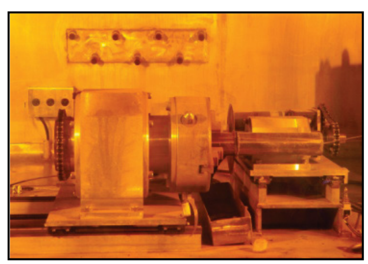
HPGe detector (backyard) for Gamma Scan

❑ M2 hot cell

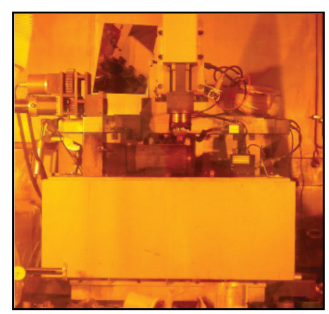
- Main Function: Capsule Dismantling, Specimen Fabrication
- Equipments: CNC milling machine, Capsule Cutting Machine, Cookie Type EDM

CNC Milling Machine
(1/1,000 mm control)





Capsule Cutting Machine

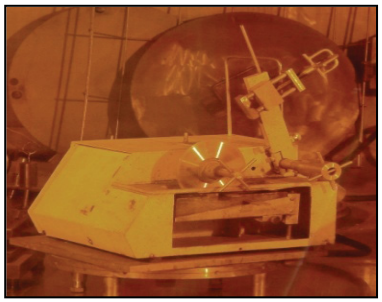


Electric Discharge Machine
(EDM)

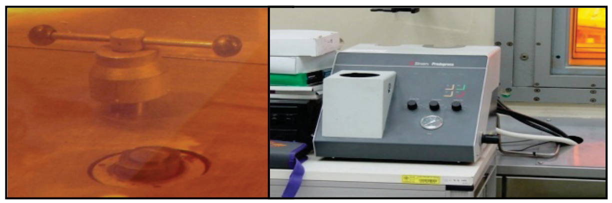
Functions in IMEF

❑ M3 hot cell

- Main Functions: Preparation of Metallographic Specimen
- Equipments: Periscope, Mounting Press, Grinder, Polisher, Cutting Saw

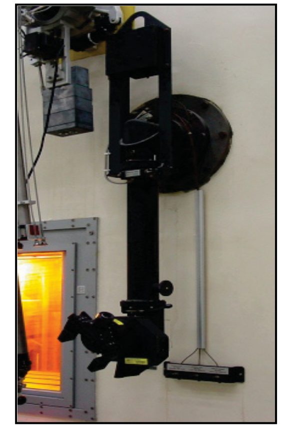


Low Speed Saw



Mounting Press

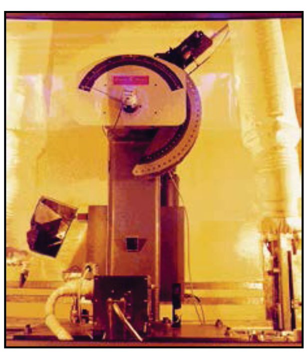
Grinder/Polisher



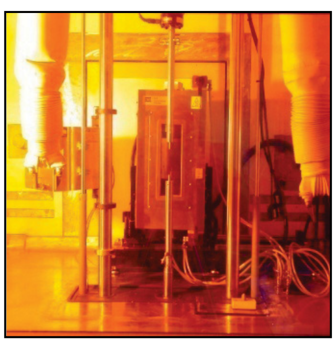
Periscope (x 20)

❑ M5 hot cell

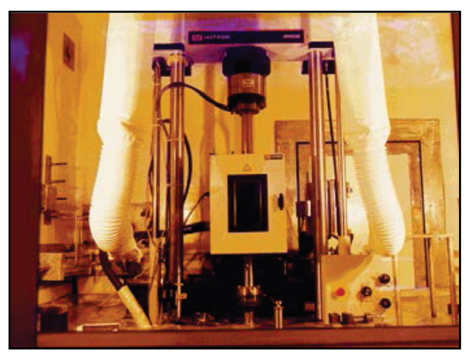
- Main Functions: Mechanical Test for Reactor Core Materials
- Equipments: Charpy Impact Tester, UTM for Small Specimen, Static UTM, Dynamic UTM



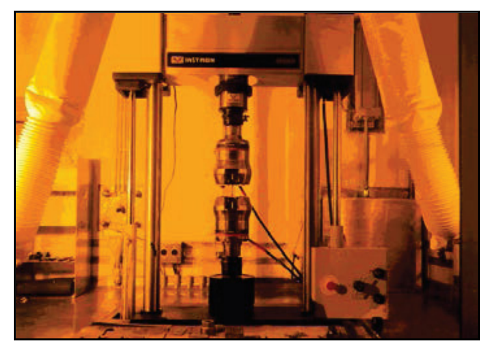
Charpy test machine



Small Specimen UTM



Dynamic Universal Testing Machine



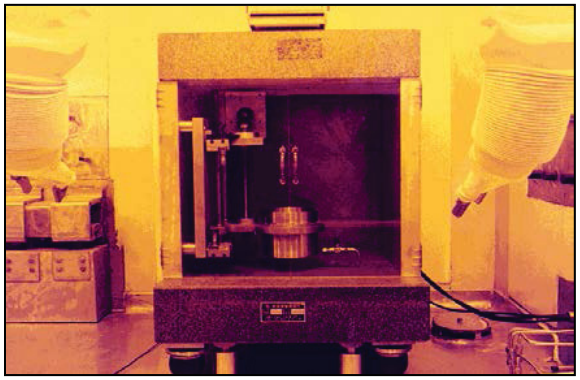
Static Universal Testing Machine

Functions in IMEF

❑ M7 hot cell

- Main Functions: Microscopic Investigation, Micro-hardness Measurement, Density Measurement.
- Equipments : Optical Microscope (LEICA Telatom3), Density Measuring Device

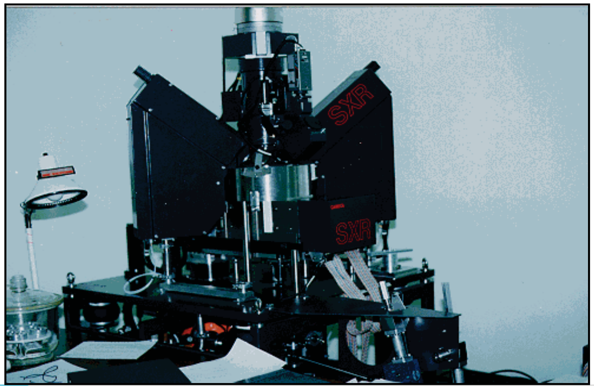
Optical microscope & Micro Hardness tester



Density measurement device (Immersion method)

❑ Hot laboratory (I)

- Main Functions: Microscopic Investigation and Chemical Composition Analysis
- Equipment: Shielded EPMA (Cameca SX-50R), TEM (Jeol)



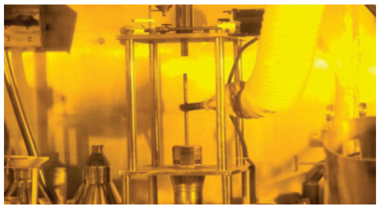
Electron Probe Micro Analyzer (WDS(WDX) -2EA)

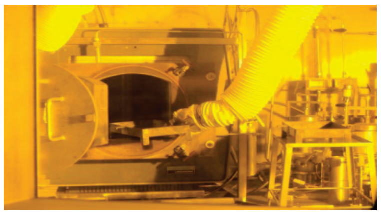
Transmission Electron Microscope

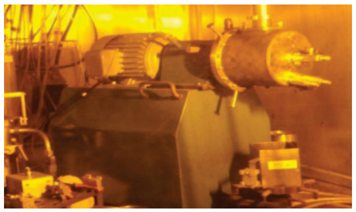
Functions in IMEF

❑ M6 hot cell

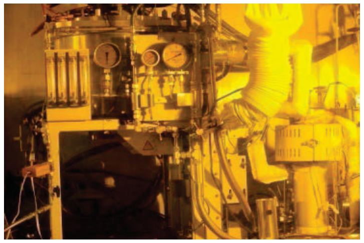
- Main Functions: Perform DUPIC Project
- Equipments: Oreox Furnace Etc


Mechanical Slitter


OREOX furnace


Horizontal rotary ball mill

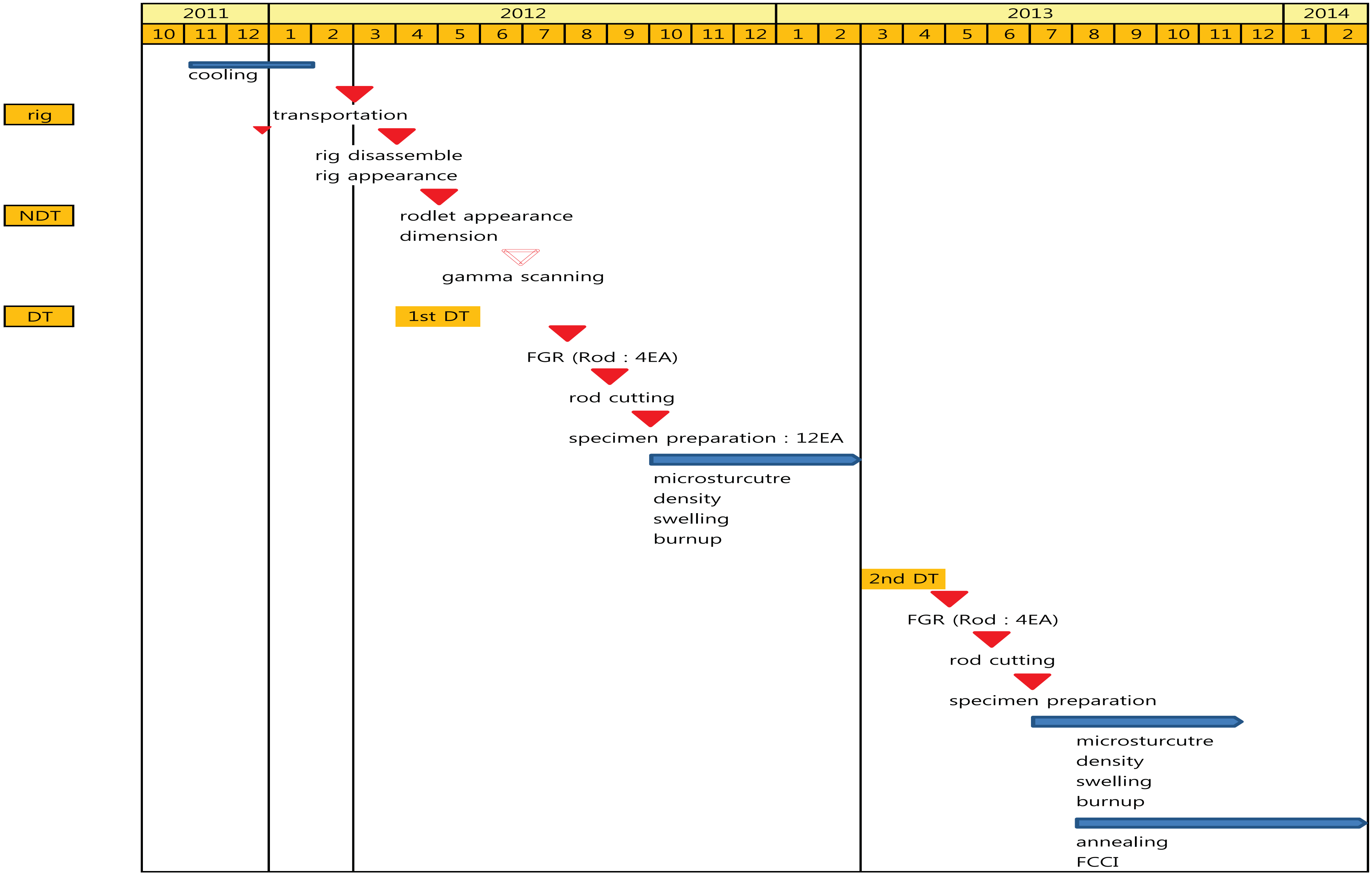

Laser Welder


Sintering furnace

❑ M8 hot cell

- Main Function: ACP (Advanced spent fuel Conditioning Process) Project
- Equipments: Slitting M/C, Vol-Oxidizer, Metallizer, Smeltzer etc

Post-irradiation examination milestone

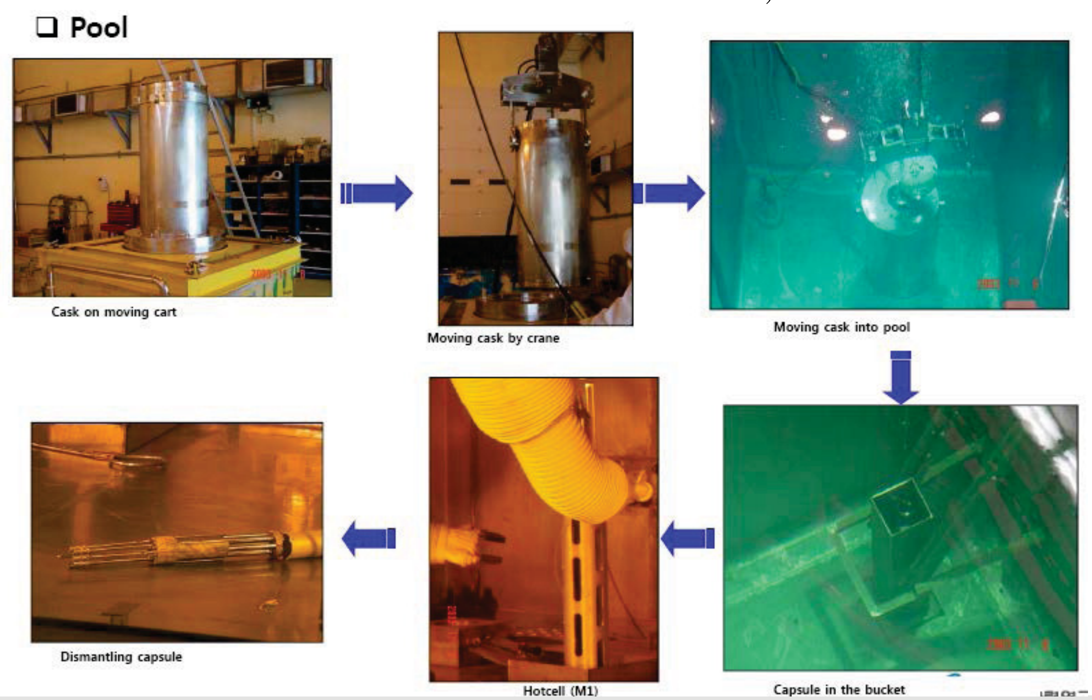


Transportation & Rig Inspection

❖ After irradiation, the capsule is transported from the HANARO pool to the IMEF and PIEF for examination

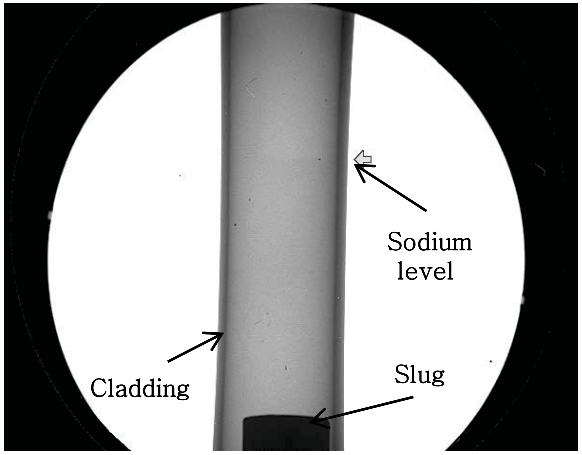
❖ Method

- Fuel transfer to hot cell
  - The fuel capsule is transferred to the IMEF through the channel connected with the pool by the use of bucket elevator
  - The fuel capsule or rods are transferred from the IMEF to the PIEF, if necessary
- Rig Inspection
  - Visual Inspection by video & digital camera
  - Disassemble by capsule cutting machine
  - ✓ Gap tightness between cladding and sealing tube

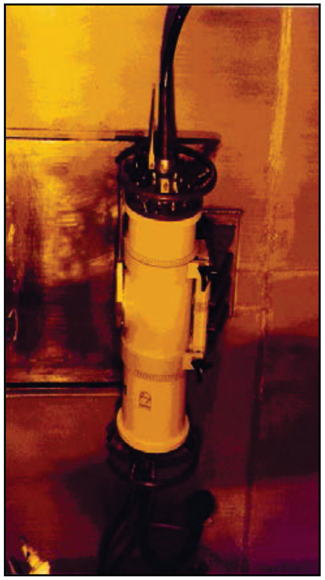


Non destructive test-Visual Inspection

- **Purpose: Integrity of fuel rodlet**
- **Application:**
  - **Sodium level and bubble**
  - **Fuel growth**
- **Equipment and Methods**
  - **Video and digital camera**
  - **X-ray radiography system**
    - **No. of rodlets : 12 EA**
- **Schedule to Complete Activity**
  - **2012. 04**
- **Issues for PIE, modeling and design**
  - **Sodium level by X-ray**



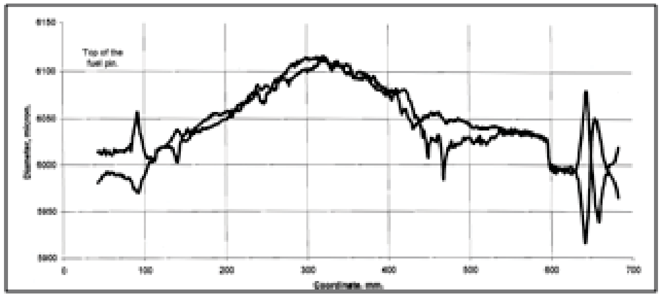
X-ray radiography



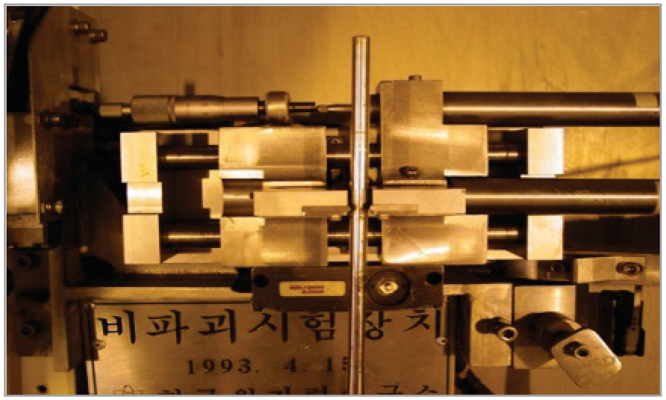
X-ray Radiography System

Non destructive test-Dimension

- ❑ **Purpose: Measurement of length and diameter of rodlet**
- ❑ **Application:**
  - ➢ **Relationship between FMS cladding strain and Fuel Slug swelling**
- ❑ **Equipment and Methods**
  - ➢ **Diameter Measurement System (LVDT)**
    - **0.5mm step, 3 times at 0, 60, 120 degree**
    - **No. of rodlets : 12 EA**
- ❑ **Schedule to Complete Activity**
  - ➢ **2012. 04 : completed**
- ❑ **Issues for PIE, modeling and design**
  - ➢ **Cladding thermal expansion and swelling according to burnup**



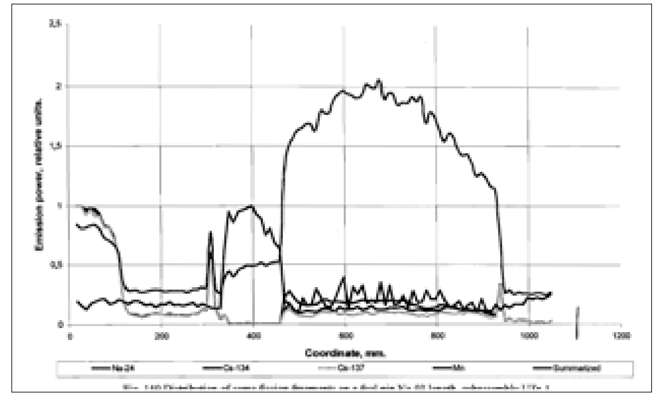
Diameter Change of Cladding



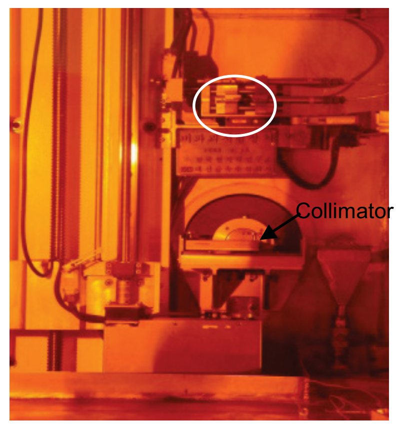
Diameter Measurement System

Non destructive test- Gamma scanning

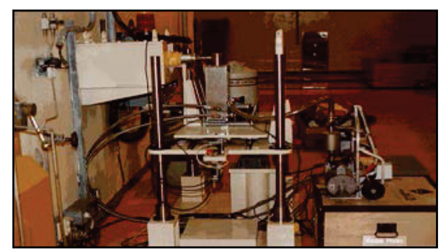
- ❑ **Purpose**
  - ➢ **Variation of burnup along axial direction of rodlet**
  - ➢ **Axial movement of fission products, especially Cs**
- ❑ **Application:**
  - ➢ **Relative fuel burnup profile**
  - ➢ **Relative distribution of various isotopes of interest in fuel**
- ❑ **Equipment and Methods**
  - ➢ **Gamma scanner in Multipurpose test bench**
    - **Vertical step travel : 2.0 mm (~80 point)**
    - **No. of rodlets : 12 EA**
- ❑ **Schedule to Complete Activity**
  - ➢ **2012. 06 : completed**
- ❑ **Issues for PIE, modeling and design**
  - ➢ **Cs distribution in Sodium**



distribution of isotopes



Gamma Scanning System



HPGe detector (backyard) for Gamma Scan

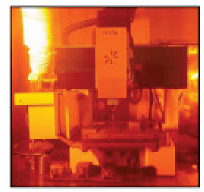
Capsule/Rodlets Disassembly and Specimen preparation

- ❑ **Purpose: Disassemble for PIE**
- ❑ **Equipment and Methods**
  - ➢ **Capsule cutting system in M2 hot cell**
  - ➢ **Preparation of specimen in M3 hot cell**
- ❑ **Schedule to Complete Activity**
  - ➢ **2012. 09 : Rodlets Disassembly completed**
- ❑ **Issues for PIE, modeling and design**
  - ➢ **Techniques for disassembling rodlets and dealing with sodium in an air cell**
    - **Steel cell & coolant**

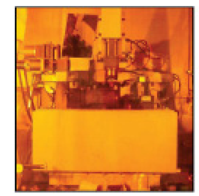
❑ M2 hot cell

- Main Function: Capsule Dismantling, Specimen Fabrication
- Equipments: CNC milling machine, Capsule Cutting Machine, Cookie Type EDM

CNC Milling Machine
(1/1,000 mm control)



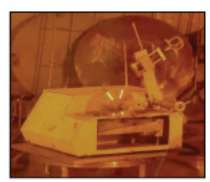
Capsule Cutting Machine



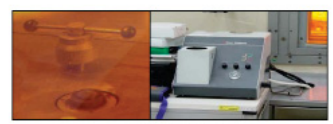
Electric Discharge
Machine (EDM)

❑ M3 hot cell

- Main Functions: Preparation of Metallographic Specimen
- Equipments: Periscope, Mounting Press, Grinder, Polisher, Cutting Saw



Low Speed Saw



Mounting Press

Grinder/Polisher

Periscope (x 20)

Rodlets Test Group for PIE

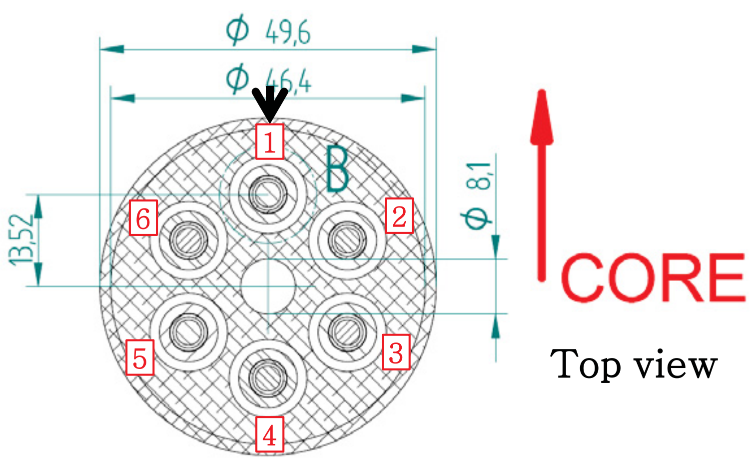
	No.	Fuel slug	Cladding	Barrier
Upper	1	U-10Zr	T92	
	2	U-10Zr	T92	
	3	U-10Zr	T92	Cr
	4	U-10Zr-5Ce	T92	
	5	U-10Zr-5Ce	T92	
	6	U-10Zr-5Ce	T92	Cr
Lower	1	U-10Zr	T92	
	2	U-10Zr	T92	
	3	U-10Zr	T92	Cr
	4	U-10Zr-5Ce	T92	
	5	U-10Zr-5Ce	T92	
	6	U-10Zr-5Ce	T92	Cr

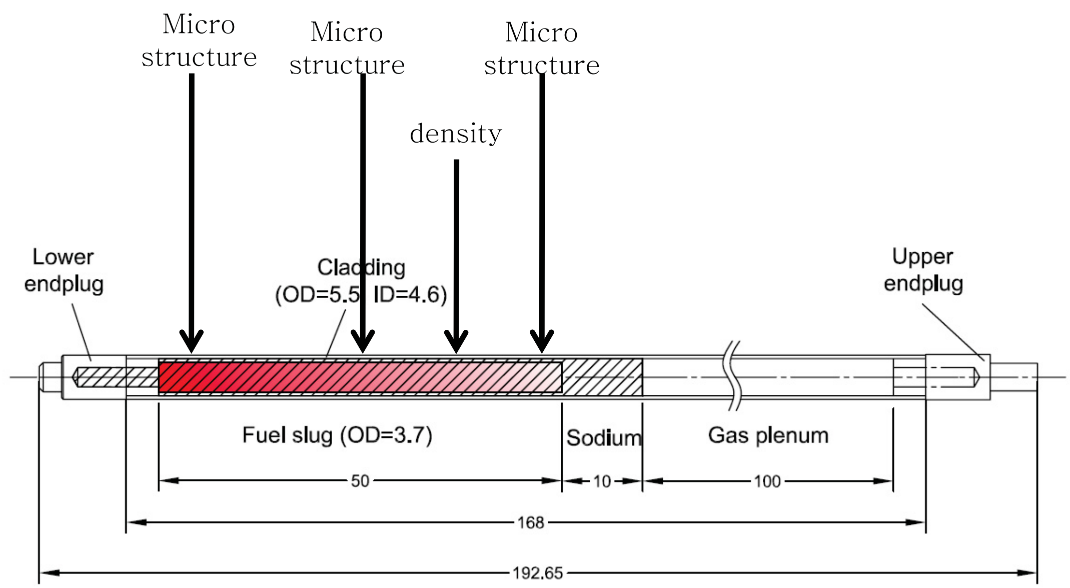
1st test

2nd test

Annealing, FCCI

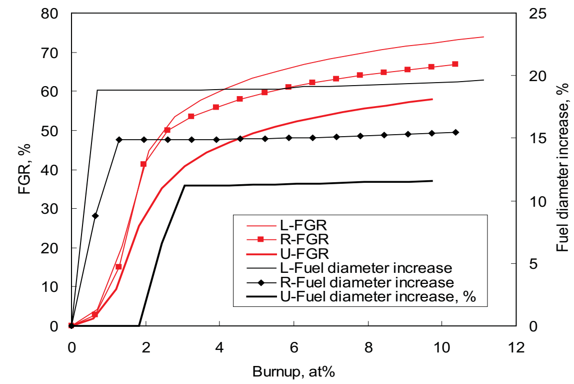
Linear power, W/cm	300
Fuel slug diameter, mm	3.7
Fuel slug length, mm	50
Sodium mass, g	0.5
Plenum volume, cc	1.82
Cr barrier thickness, μm	20
Hf thickness, mm	1.4 & 1.6



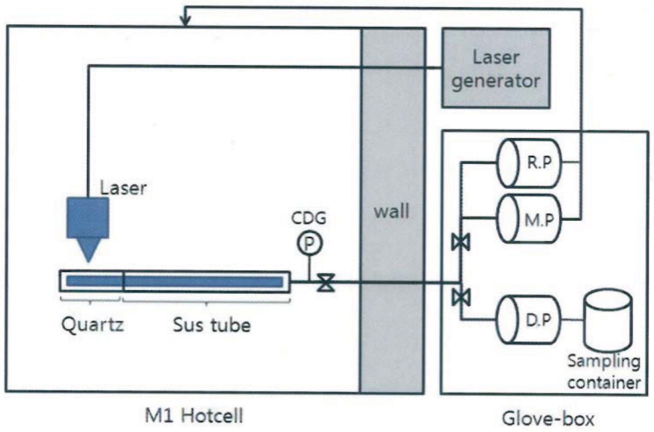


Destructive test- Fission gas release

- ❑ **Purpose: Variation of FGR with fuel burnup**
- ❑ **Application:**
  - ➢ **Determine fission gas release**
- ❑ **Equipment and Methods**
  - ➢ **Puncture system**
    - • **System only for SFR metal fuel needed**
  - ➢ **No of specimen :**
    - • **1st : 4EA, 2nd : 4EA**
- ❑ **Schedule to Complete Activity**
  - ➢ **2012. 07 : 1st test completed**
- ❑ **Issues for PIE, modeling and design**
  - ➢ **Uncertainty in measurement – small system needed**



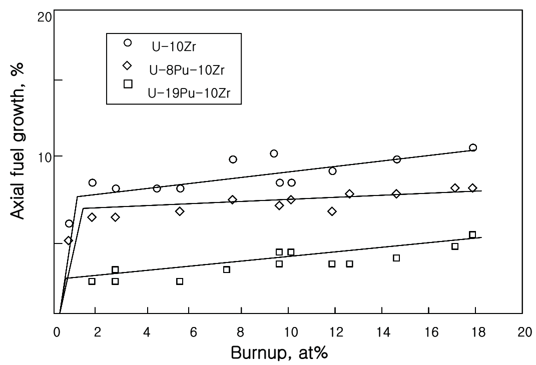
Fission gas release

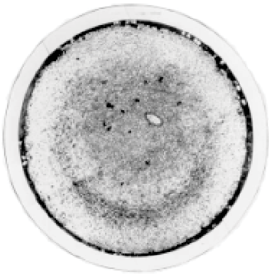


puncture system

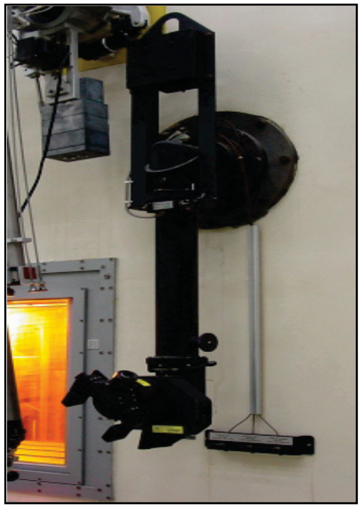
Destructive test-Swelling and axial length

- **Purpose: Measurement of variations in swelling and axial length with fuel burnup**
- **Application:**
  - **Relationship between Fuel irradiation growth and swelling**
- **Equipment and Methods**
  - **Measure by Highscope and OM**
    - **Compare with density**
  - **No of specimen**
    - **1st : 12EA, 2nd : 12EA**
- **Schedule to Complete Activity**
  - **2012. 08 : 1st test completed**
- **Issues for PIE, modeling and design**
  - **Measurement according to indirect measurement**





irradiation growth and swelling



Periscope (x 20)

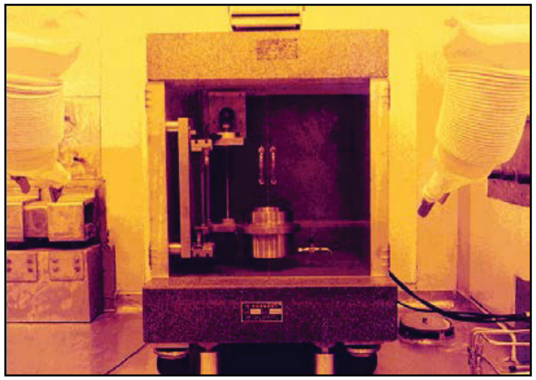
Optical microscope & Micro Hardness tester

Destructive test-Density

- Purpose: Measurement of density variation
- Application:
  - Quantify irradiation swelling
  - Derive thermal conductivity
- Equipment and Methods
  - Immersion method
    - Micro-Balance & toluene
  - No of specimen
    - 1st : 4EA, 2nd : 4EA
- Schedule to Complete Activity
  - 2012. 08 : 1st test completed
- Issues for PIE, modeling and design
  - Measurement considering the porosity of irradiated metal fuel

Parameter	Value	
	UZr-1	UZr-2
Mean density, g/cm^3	15.17	15.365
Minimum density, g/cm^3	13.68 ?	15.21
Maximum density, g/cm^3	15.45	15.71
Standard deviation, g/cm^3	0.102	0.423

The standard deviation was defined by the formula: $\{[n\Sigma x^2-(\Sigma x)^2]/n^2\}^{1/2}$.

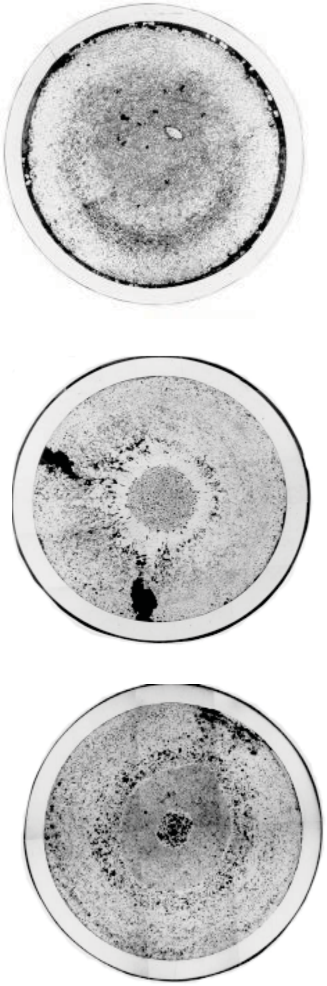


Density measurement device
(Immersion method)

Destructive test-Microstructure

- **Purpose: Size and distribution of porosity, distribution of phase and fission product, Infiltration of sodium into fuel**
- **Application:**
  - **Fuel phase and porosity, Fuel constituent migration, sodium infiltration, FCCI etc**
- **Equipment and Methods**
  - **Optical Microscope (LEICA Telatom3)**
  - **Mini-SEM**
  - **Shielded EPMA**
  - **No of specimen**
    - **1st : 12EA, 2nd : 12EA**
- **Schedule to Complete Activity**
  - **2013. 02 : 1st test completed**
- **Issues for PIE, modeling and design**
  - **Sample preparation**

Optical microscope & Micro Hardness tester



Destructive test- Burnup analysis

- **Purpose: Measurement of fuel burnup with chemical analysis**
- **Application:**
  - **Derive burnup**
- **Equipment and Methods**
  - **The burnup is determined by the measurement of 148Nd separated chemically from the fuel sample**
  - **by means of mass spectrometry such as LA-ICP-MS(Laser Ablation Inductively Coupled Plasma Mass Spectrometry).**
  - **Burnup measurement is performed in the Chemical Lab. of the PIEF.**
  - **The samples to be analyzed are transported by a small cask from the IMEF to the Chemical Lab. of the PIEF.**
- **Schedule to Complete Activity**
  - **2013. 02 : 1st test completed**
- **Issues for PIE, modeling and design**
  - **Standard data in Fast Reactor Conditions**

LA-ICP-MS

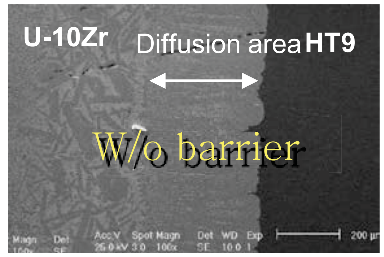
Diffusion couple test between fuel and barrier cladding

❖ Status

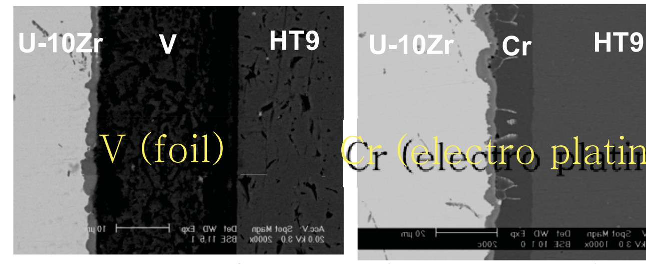
- ❖ KAERI fabricated the barrier cladding
- ❖ Diffusion couple testing against metal alloys were carried out.
  - ❖ Barrier(Zr, V metal foil, Cr) and rare earth element(Misch metal, Nd) interaction
- ❖ The performance of Barrier such as V & CrN showed a good result.
- ❖ Barrier cladding tubes by electroplating of Cr Under irradiation at HANARO (2010. 11.)

❖ Planned Activities

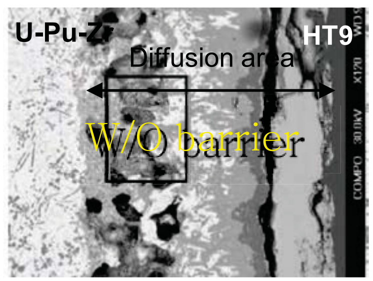
- ❖ Focuses on the fabrication of barrier cladding tubes
- ❖ Cr plating, Ion Nitride, and combining Cr plating/Nitride Barrier
- ❖ ATR irradiation through Collaboration with INL



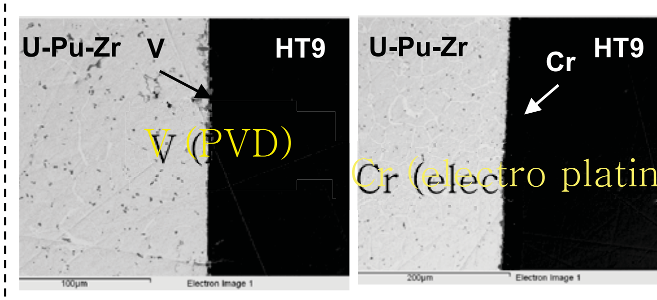
U-Zr /HT9



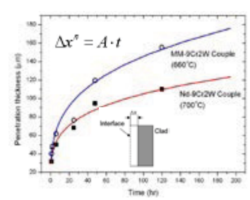
U-Zr/barrier (800°C, 25hrs)

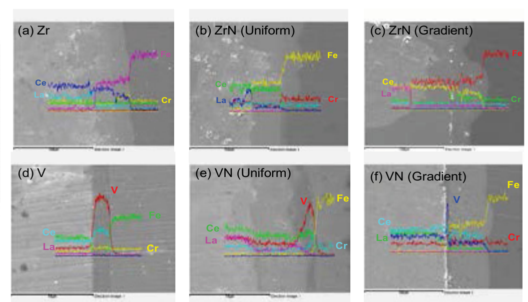


U-Pu-Zr/HT9

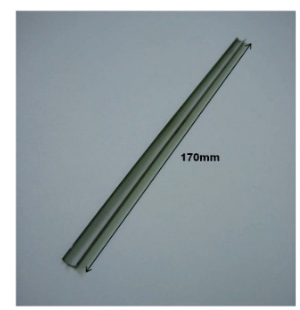


U-30Pu-20Zr/barrier (750°C, 24hrs)





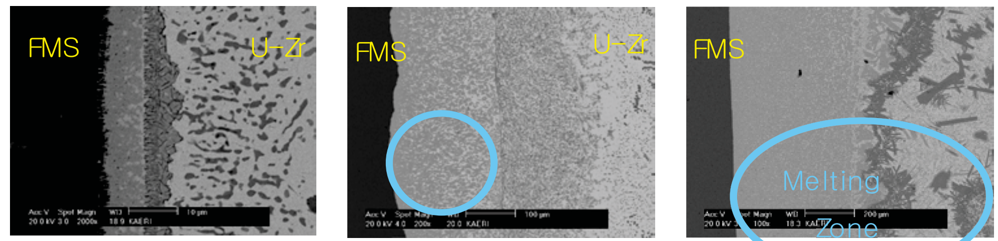
Diffusion couple test of RE/cladding/barrier material



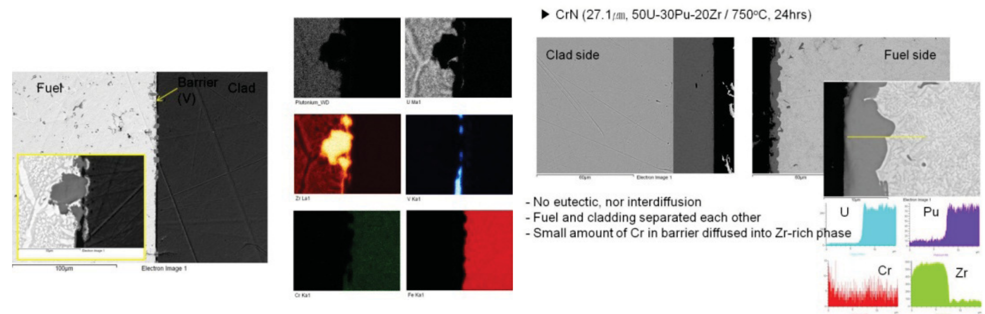
Cr Barrier tube for Irradiation Test

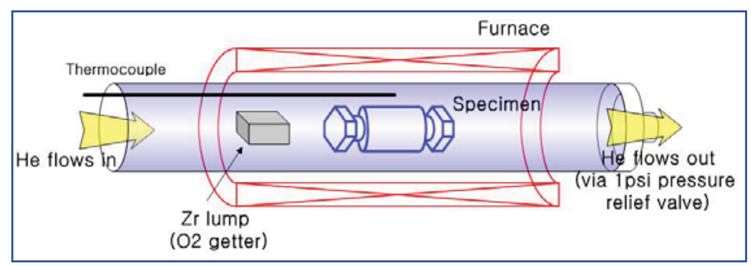
Destructive test-FCCI test

- **Purpose: Fuel-cladding chemical interaction test with irradiated fuel**
- **Application:**
  - **Evaluate FCCI behavior**
- **Equipment and Methods**
  - **Diffusion couple test in shielded globe box**
- **Schedule to Begin Activity**
  - **2013. 07**
- **Issues for PIE, modeling and design**
  - **Upgrade of Diffusion couple methods**
    - **Design and fabrication of device for diffusion couple test**



700°C→ Diffusion layer 740°C→ Eutectic 800°C→ Eutectic

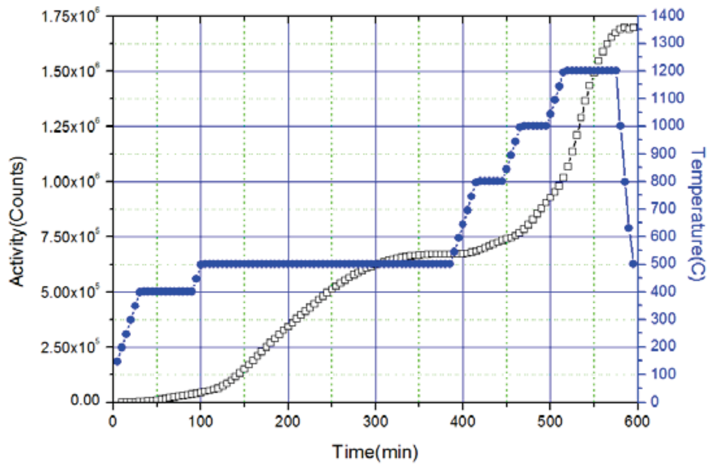




FCCI test with irradiated fuel

Destructive test- Annealing test

- **Purpose: Behaviors of swelling and fission gas release during simulated transient tests with irradiated fuel**
- **Application:**
  - **check the fission gas behaviors during transient state.**
- **Equipment and Methods**
  - **Annealing furnace**
  - **heated up to the high temperature range (less than 1000℃)**
- **Schedule to Begin Activity**
  - **2013. 07**
- **Issues for PIE, modeling and design**
  - **Measurement of swelling behavior related to fission gas bubble behavior**



Outstanding results in IMEF and PIEF

Origin	Item	Duration (yr)	Purpose	Remark
Fuels for National R&D Projects	Advanced Research Reactor Fuels	2001 ~ Cont'd	• Evaluation of the atomized U-Mo fuels (4 phases) • Reaction layers, swelling, burn-up measurements etc	
	HANARO Fuel Assemblies	1998~ 2003	• Evaluation of the dispersed U-Si fuels (3 assemblies) • Burn-up distributions, shape deformations etc	
	DUPIC Fuel Rigs	2003~ 2007	• Analysis of the irradiation behaviors (6 phases) • Burn-up, shape deformation, fuel distributions etc	
	SMART Fuels	2002~ 2007	• Evaluation of the SMART U-Zr fuels (3 phases) • Burn-up, composition distribution, blistering etc	
	Double Cooled Annular Fuels	2009 ~ Cont'd	• Analysis of the advanced fuel meat shape fuels • Dimension, burn-up, composition distribution etc	
Fuels for Power Plants	Defected Fuels from all PWRs in Korea	1987 ~ Cont'd	• Cause analysis of the defective fuels (10 assemblies) • Damaged surface investigation, non-destructive etc	PIEF
	Advanced PWR Fuels	2004 ~ Cont'd	• In-reactor behavior analysis ACE-7, PLUS-7 FA's • Burn-up, cladding integrity, dimensional change etc	PIEF
Materials for National R&D Projects	Gen-IV Reactor Core & Structural Materials	2001 ~ Cont'd	• Analysis of the future reactor materials (6 times) • Tensile, creep, facture surface analysis etc	
	New PWR Vessel Materials	2007 ~ Cont'd	• Analysis of the future PWR vessel materials (4 times) • Tensile, fracture, impact etc	
	PWR Fuel Skeletons	2008 ~ Cont'd	• Behavior evaluation of the skeleton parts (3 skeletons) • Mechanical tests for nozzle, control rod, cladding etc	
	SMART S/G Tube Materials	2009 ~ Cont'd	• Irradiation evaluation on the tube materials (3 capsules) • Tensile, fracture, thermal conductivity etc	
Materials for Power Plants	Reactor Vessel Surveillance Program	1998 ~ Cont'd	• Reactor vessel aging managements (16 reactors) • Impact, tensile, chemical composition analysis etc	
	Life Extension Assessment of Kori-1 Reactors	2003~ 2006	• Integrity evaluations of reactor vessel weldments • Impact, fracture etc	
	CANDU Pressure Tube Aging	2001~ 2005	• Evaluation of the operated pressure tubes (2 tubes) • Tensile, fracture, DHCV etc	

Future PIE plan of IMEF

Origin	Group	2010	2011	2012	2013 ~ 2015 (03 yrs)	2016 ~ 2019 (04 yrs)
National R&D Projects	Fuels	KOMO-IV Fuels	SFR-I Fuels	Double Cooled-II Fuels	VHTR-I Fuels; FTL Irrad. Fuels	SFR-II Fuels; VHTR-II Fuels
	Materials	SMART S/G Tube Materials	PWR FA Parts		HANARO Reflector Surveillance; SFR Reactor Materials	VHTR Carbon Structure; ITER Structural Parts
Power Plants	Fuels	YGN4 Defective Fuel	ACE7 LTA Fuel Rods		Plus7™ Fuels	HIPER Demonstration Fuel
		Plus7 Skeleton	ACE7 LTA Fuel Rod/ Skeleton		KNF Developed Fuel Skeleton; HANA Clad Fuel	
	Materials	YGN 6-1st Surveillance	UJ 1-6th Surveillance	UJ 2-6th Surveillance	YGN 3-2nd Surveillance; UJ 3-2nd Surveillance	UJ 4-2nd Surveillance; UJ 5-1st Surveillance; UJ 6-1st Surveillance
		CRUD Analysis	Storage FA Skeleton		Wolsong-2 PT Surveillance; Damaged Structure Parts from PWR	Wolsong-3, 4 PT Surveillance
Development of PIE Techniques		Thermal Conductivity	VHTR TRISO Fuel		FTL Dismantling & Test Techniques	Sodium Bonded SFR Fuel
		Installation of New Shielded EPMA		CANDU DHCV Test	Small Volume Rod Puncturing	Aqueous SCC Test
Technical Support		Utility Support for DUPIC and ACP Projects (1998 yr ~)				
		Basic Research Support for Universities (1994 yr ~)				

4

CONCLUSIONS

Conclusions

- Metallic fuel development for SFR started in 2007
- Irradiation in HANARO : 2010.11
- PIE will be carried out in IMEF
  - Totally 8 hot cells with 72 m in a hot cell length, 1 pool, 2 hot lab's and 31 working units (31 windows)
- PIE of SFR metal fuel
  - PIE considering Na treatment
  - System for FGR measurement
  - In-reactor Behavior only for SFR metal fuel such as FCCI, Fuel constituent migration etc
- Continuous experimental tests needed

  ➔ for verifying the performance of metal fuel

  ➔ International collaborations are essential

U.S. DEPARTMENT OF
ENERGY
Nuclear Energy

United States Perspective on Post-Irradiation Examination Capabilities

OECD/NEA International Workshop

Dennis Miotla
Deputy Assistant Secretary for
Nuclear Facility Operations
U.S. Department of Energy

June 16, 2011

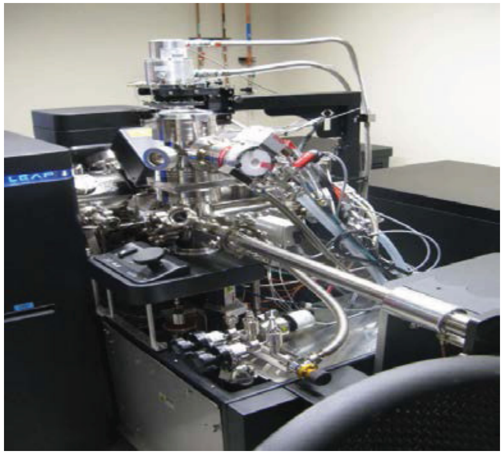
So, Why Am I Here Representing the U.S. DOE at this Workshop?

- We are in the stage of identifying what capabilities are needed; what are the gaps in the United States (U.S.) and international research communities
- We want to complement, not compete, with capabilities that already exist or are planned world-wide
- We plan to invest in PIE capabilities and facilities that are:
  - adaptable to changing technologies,
  - sustainable, given the constant demand on limited resources, *and*
  - accessible to researchers world-wide

DOE-Office of Nuclear Energy is working to establish an enduring capability for nuclear energy research in the U.S.

Advancements in PIE Capabilities

- During the past 15 years, stunning advancements in analytical research instrumentation have been made
- Nano-scale (10^{-9} meter) characterization of materials is becoming routine, with capabilities for sub-angstrom (10^{-10} meter) investigation increasing
- Examples of new capabilities of interest to Department of Energy include:
  - **Nano-indenter** (have)
  - **Laser Resonant Ultrasonic Spectroscopy**
  - **Dilatometer**
  - **Scanning Thermal Diffusivity Microscope**
  - **Analytical Transmission Electron Microscope** (have)
  - **Dual Beam Focused Ion Beam** (have)
  - **Local Electron Atom Probe** (have)



Atom Probe

Overall U.S. DOE-Office of Nuclear Energy Strategy for Nuclear Research and Development

- Concentration on restoring the U.S. nuclear energy research capability to world-class status
- **Consolidate and maximize existing capabilities wherever they may exist**
- **Co-locate new capabilities with existing infrastructure**
- Increase international collaboration and make greater use of university capabilities
- Focus on the core capabilities, such as **hot cell facilities**, and specialized facilities needed to conduct **post-irradiation examination and related mechanical testing**

Reference: Facilities for the Future of Nuclear Energy Research, A Twenty-Year Outlook (Feb. 2009)

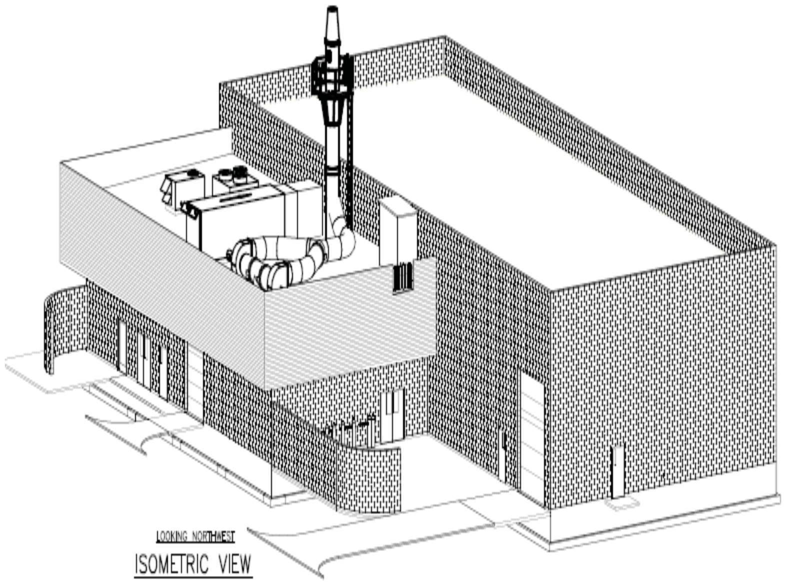
Examples of Existing PIE Capabilities at U.S. National Laboratories

U.S. Location	PIE Capabilities
Idaho National Laboratory	• On-site irradiation capability at the Advanced Test Reactor • Large, heavily shielded hot cells capable of handling full-size fuel assemblies and elements • Analytical laboratories with fuel examination capabilities • Specialized PIE equipment
Los Alamos National Laboratory	• Hot cells with ability to accommodate research quantities of materials • Analytical laboratory capabilities
Oak Ridge National Laboratory	• On-site irradiation capability at the High Flux Isotope Reactor • Hot cells with ability to accommodate research quantities of materials • Analytical capabilities for material characterization

Current U.S. Activities for PIE (1-3 Years)

- **Re-baselined PIE capabilities to ensure linkages to Research and Development needs**
- **Procured state of the art PIE equipment to support missions**
- **Constructing Irradiated Materials Characterization Laboratory to house state-of- the art PIE/analytical equipment with environmental control**
- **Refurbishing hot cells and supporting infrastructure**

Irradiated Materials Characterization Laboratory



Goal: By 2013 provide comprehensive PIE capabilities to match current world status

What Have We Done so Far to Identify Future Capabilities and Needs for Advanced PIE? (5-10 Years)

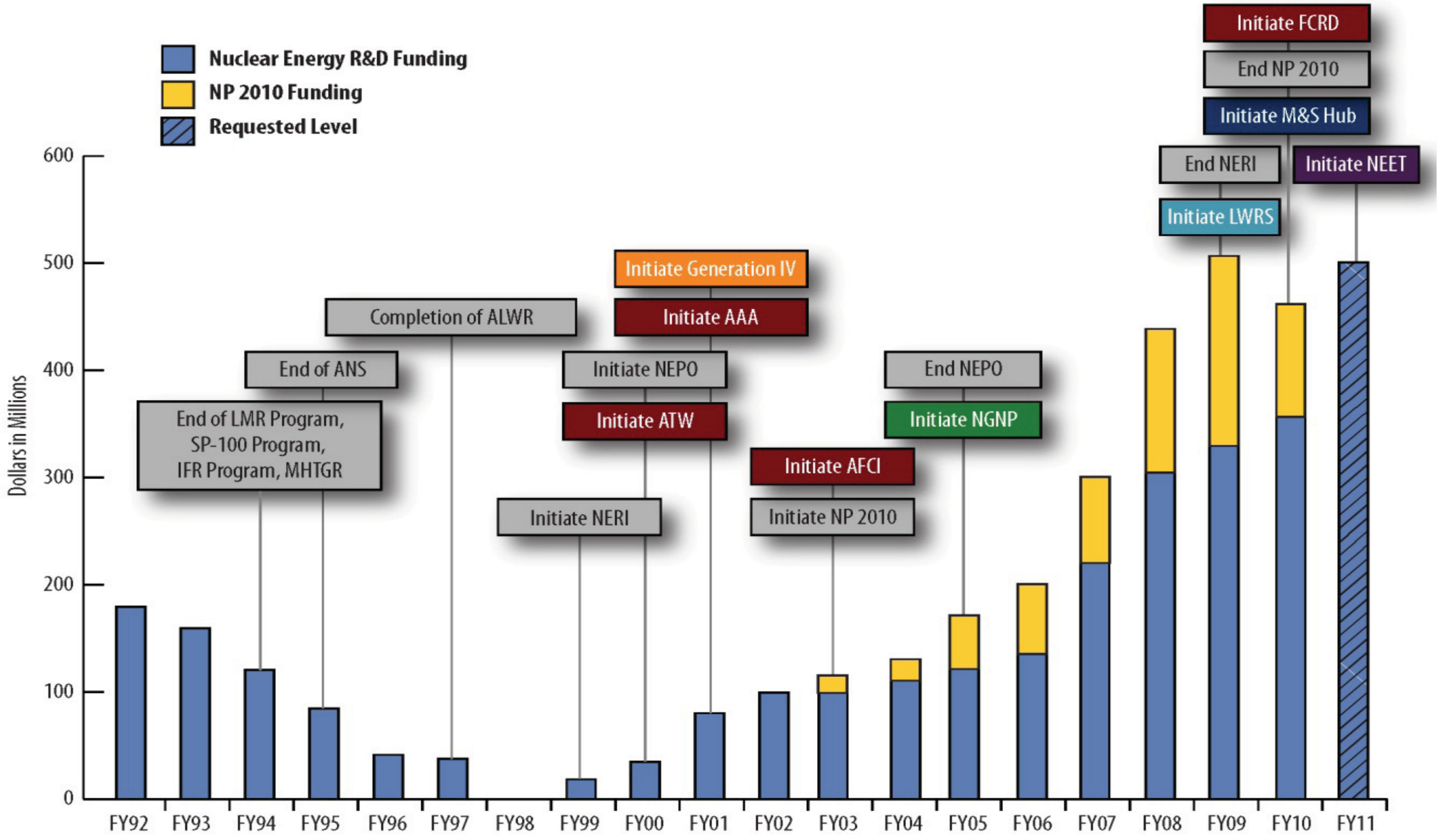
- Formally identified the gap in long-term PIE needs in January 2011
- Requested funds to conduct exploratory studies to assess possible advanced PIE capability options
- Held a United States PIE Workshop March 29-30, 2011, in Gaithersburg, Maryland, to identify the domestic needs for PIE to support advanced fuels and nuclear material development.
  - *Conclusions of workshop will be used to identify alternatives*
  - *National PIE needs were identified from various perspectives, including universities, industry, vendors, Nuclear Regulatory Commission (NRC), Department of Energy (DOE), and DOE national laboratories*

We are interested in knowing the international perspective

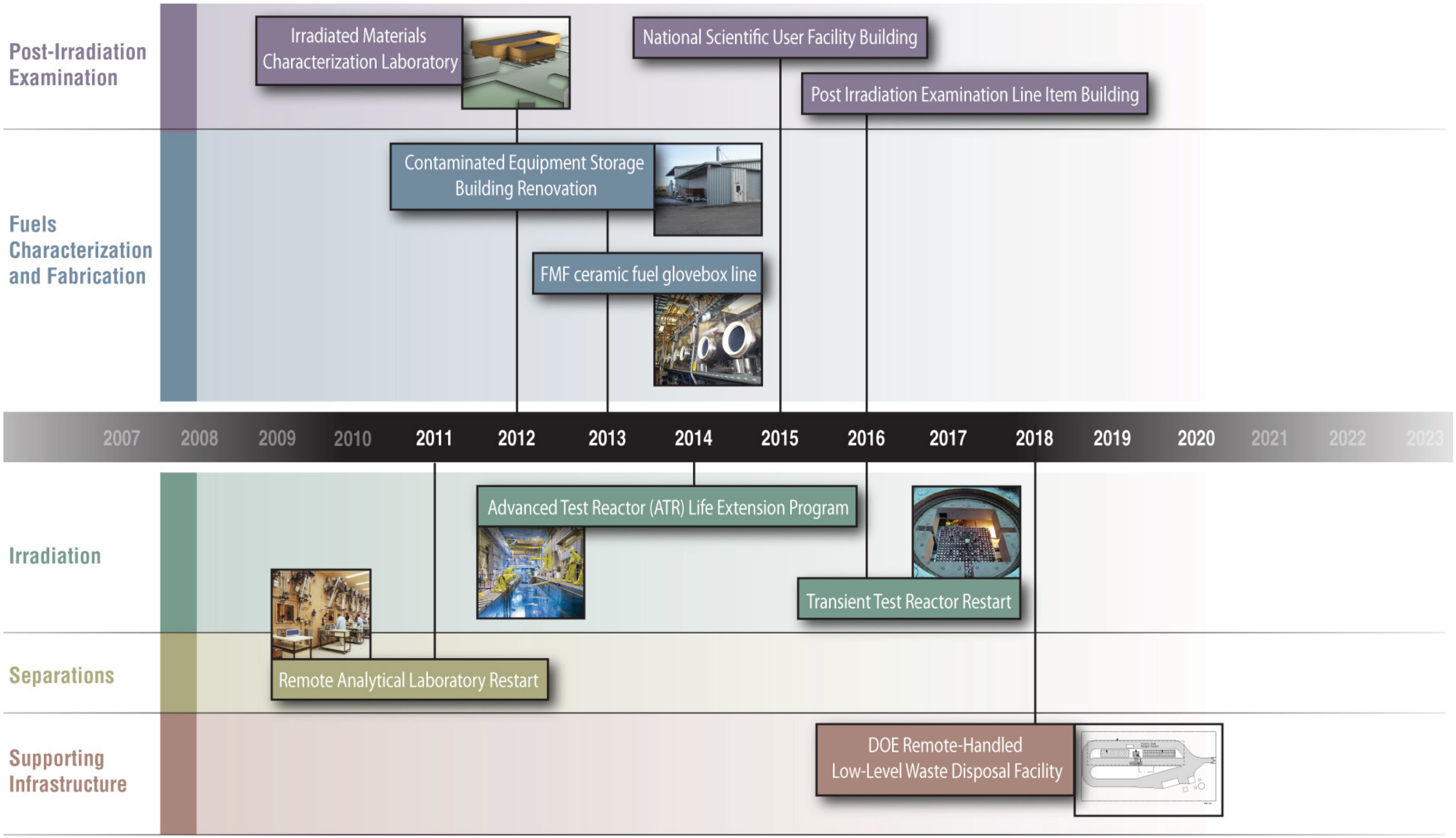
U.S. DEPARTMENT OF ENERGY

Nuclear Energy

Historical Funding Trend for Nuclear Energy Research and Development Activities



Proposed Timeline for Establishing New Core and Enabling Capabilities at the Idaho National Laboratory



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Summary and Conclusion

- United States is engaging in PIE capabilities and is investing in near-term activities to complement the existing international suite

- Interested in looking toward the next generation of PIE capabilities to make smart investment decisions

Questions?

Fuel performance code :

characterization and PIE needs for modeling validation.

V. Bouineau,

CEA-Cadarache, Fuel studies department

Simulation: main issues

- Interpretations of experiments conducted on nuclear fuel:
  - *Out of pile experiments*
  - *In pile experiments*

- Design of fuel elements:
  - *Fuel behavior improvements*
  - *Innovative designs (fuel or nuclear systems)*

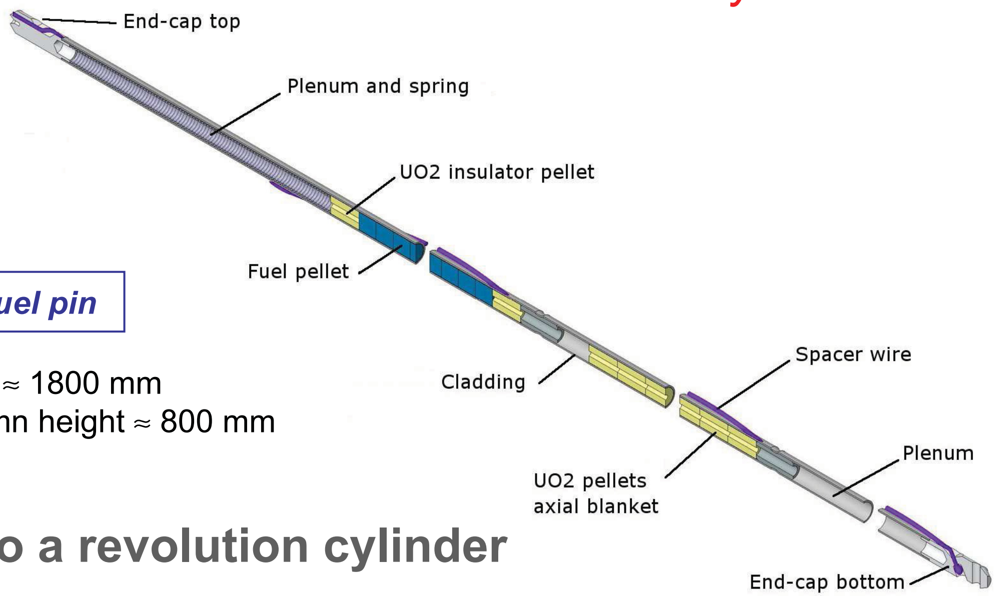
- Design of experimental studies on irradiated fuels:
  - *Irradiation in SFR (Phenix, …) and MTRs*
  - *Out of pile experiments*

- Capitalization of acquired knowledge:
  - *Fuel Performance code like Germinal (SFR)*

Modeling: main challenges

Physics and modeling to describe the behavior of fuel pins with mixed oxide fuel $(U,Pu)O_2$

Thermal analysis *Mechanics* *Fuel Physics and Chemistry*



PHÉNIX fuel pin

Total height ≈ 1800 mm
Fissile column height ≈ 800 mm

Geometry assimilated to a revolution cylinder

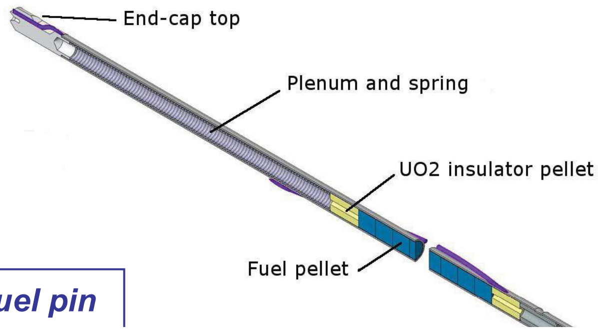
Axisymetrical model 1,5D :

- ✓ **Radial resolution coupling axial slices by thermalhydraulics in coolant**
- ✓ **Similar as the model for PWR fuel rods**
  ⇒ Benefits of recent work on PWR fuel rods simulation

Modeling: main challenges

Physics and modeling to describe the behavior of fuel pins with mixed oxide fuel $(U,Pu)O_2$

Thermal analysis *Mechanics* *Fuel Physics and Chemistry*



PHÉNIX fuel pin

- **Fuel element structural integrity**

Structural computation

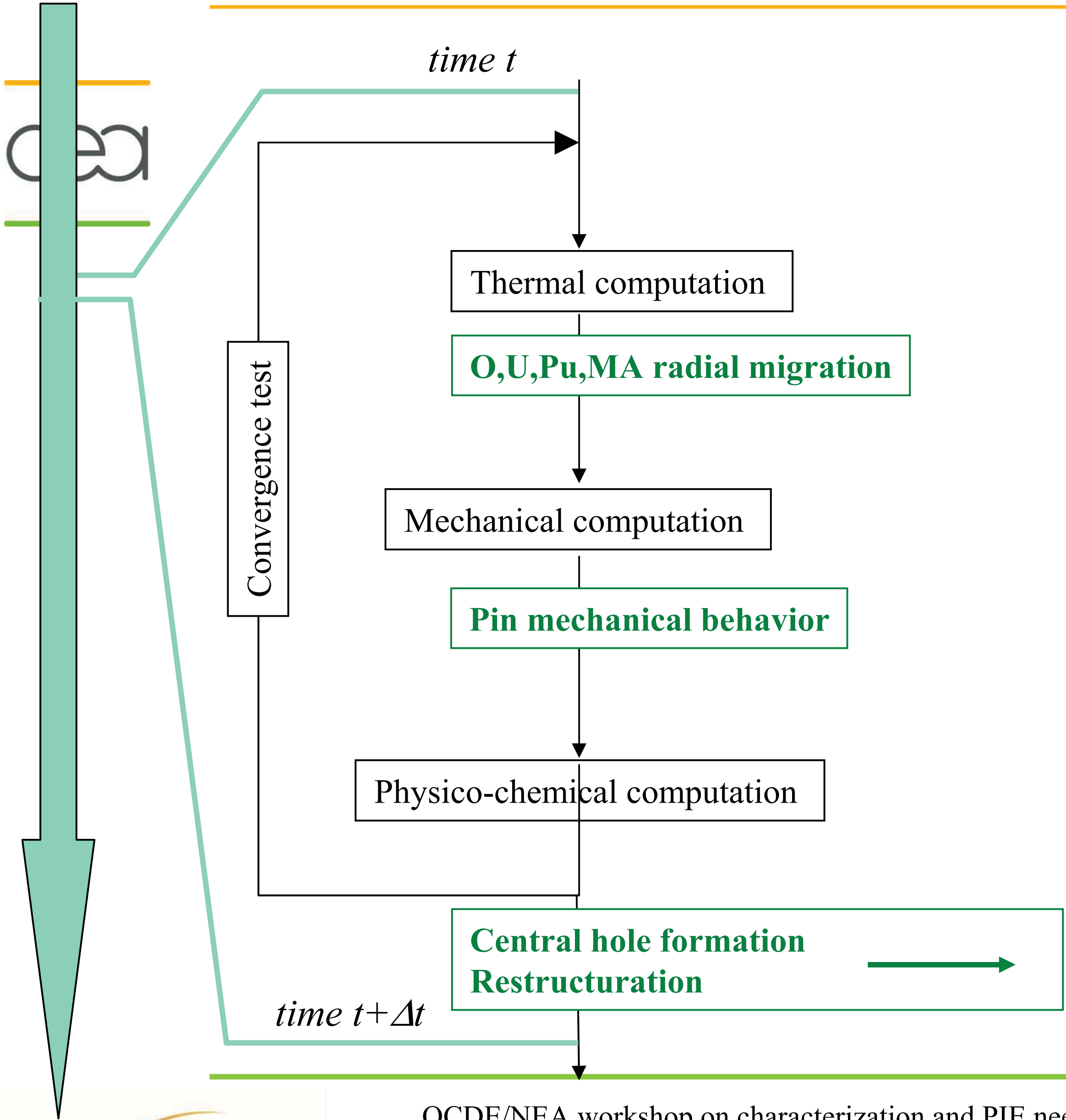
Thermo-mechanical state of the fuel element

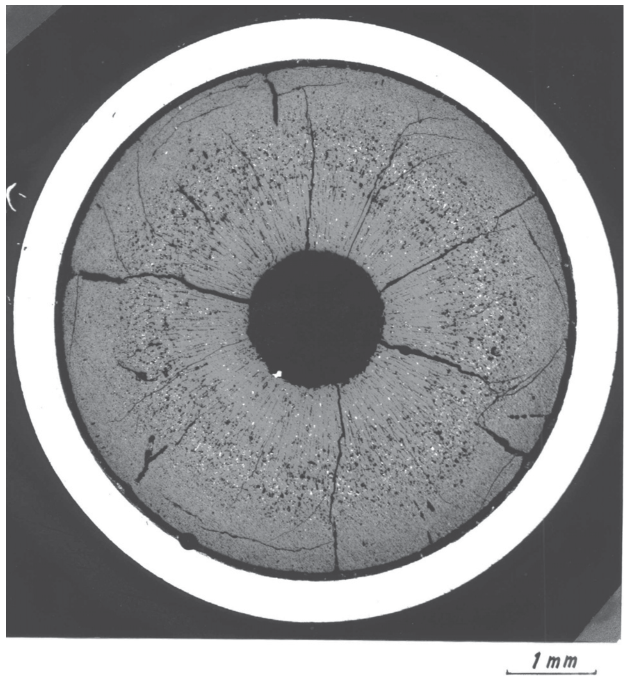
- **Fuel behavior**

Physico-chemical of irradiation

Inventory and localization of fission products in the pellet

GERMINAL V2


time t
Thermal computation
O,U,Pu,MA radial migration
Mechanical computation
Pin mechanical behavior
Physico-chemical computation
Convergence test
Central hole formation
Restructuration
time t+Δt


1 mm

Synthesis of couplings

Fuel and cladding deformations modify the gap thickness and consequently heat exchange.

Thermal analysis

Mechanics

Fuel and cladding mechanical properties depend on temperature. Thermal creep of fuel and cladding.

Thermal activation of physics

Fuel thermal properties depend on (O/M), (Pu/M), porosity.

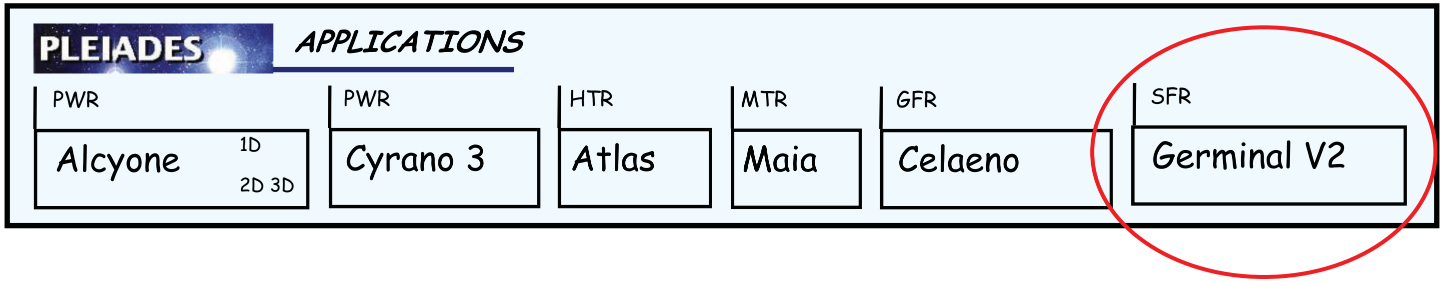
Evolution of gap state (JOG), and consequently of heat exchange between fuel and cladding.

Radial migration of oxygen
Radial migration of porosities / Pu
Creation & release of fission gases / Cs

Fuel mechanical properties depend on (O/M) and porosity.

Fuel swelling is modified by porosity migration and fission products release.

GERMINAL in PLEIADES



- Federative unified framework to develop fuel performance codes

  Co-developed by CEA and eDF

  Allows modeling advancement to be shared among all applications

  Involves **connexions with Data Bases** :

  ✓ **Physical Data Base** : **Sirius** ⇒ Material properties

  ✓ **Validation Data Bases** : CRACO (PWR rods), **BREF** (SFR pins)

Benefit of PLEIADES platform : Inheritance of mechanical behaviors modeling of PWR fuel rods materials

Fuels with minor actinide

- Homogeneous mode : 1-5% MA in mixed oxide fuel $(U,Pu)O_2$
  - In core with high temperature

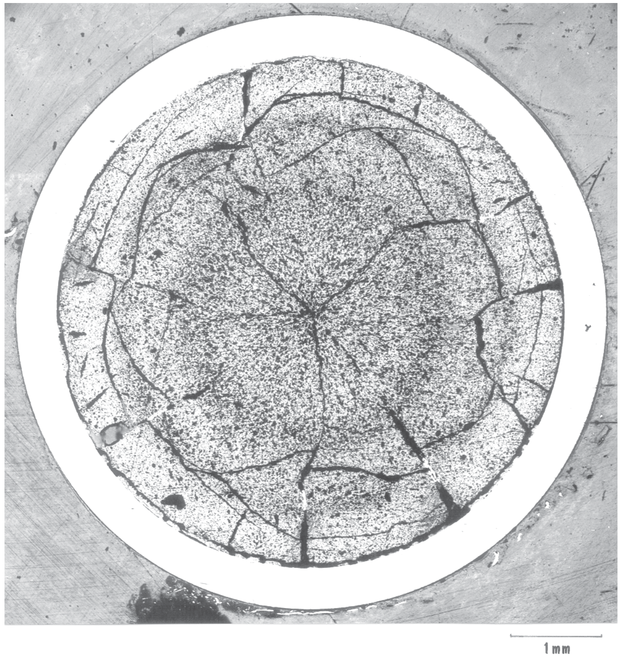
- Heterogeneous mode : 10-20% MA in uranium oxide fuel
  - At the periphery of the core with low temperature

- On Inert Matrix : 10-20% MA on typically MgO
  - At the periphery of the core with low temperature

  - **Main differences**
    - Higher production of Helium
    - Modification of physical properties (oxygen potential, thermal conductivity, temperature of fusion, …)

Experimental results on Am transmutation

- **SUPERFACT-1 in PHENIX (1988)**



- **Heterogeneous pin U,Am(20%),Np(20%)O_2 up to 4 at%**
- **Cladding-fuel mechanical interaction (cladding strain + circumferential cracks)**
- **No classical restructuration but large central region with porosity**

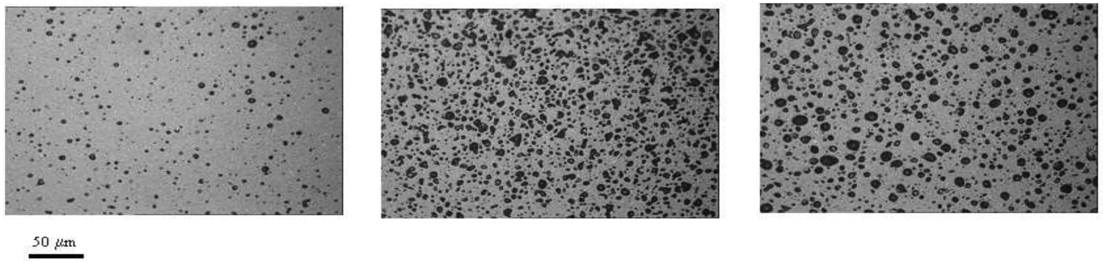
→ No correct fuel properties in actual code

- **Experimental data are capitalized in database BREF (fabrication, irradiation and PIE)**

Experimental results on Am transmutation

- **Experiment in inert matrix**
    - EFTTRA T4 and T4bis in HFR

Edge | Am-rich zone | Pellet Centre



Area porosity:

~5% | ~32% | ~25%

- Large swelling of 18 to 24%
- Large helium retention (80%)
- Up to 32% of porosity locally

Specific development for Fuels with minor actinide

- Homogeneous mode : 1-4% MA in mixed oxide fuel $(U,Pu)O_2$
  - In core **with high temperature**
    - » Helium release
    - » Modification of Oxygen Potential
    - » Modification of O, Pu, MA Redistribution
    - » Impact on thermal conductivity
    - » Impact on cladding internal corrosion
    - » Impact on melting temperature

- Needs for physical properties on fresh and irradiated fuels
  - » **Feed the Physical Data Base** : **Sirius**

Specific development for Fuels with minor actinide

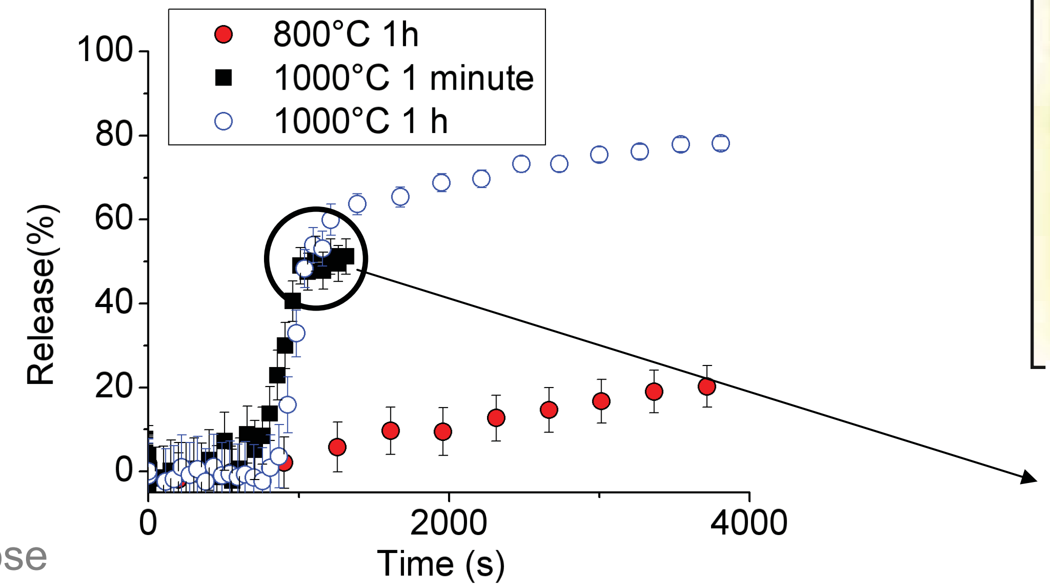
- Heterogeneous mode : 10-20% MA in uranium oxide fuel
- On Inert Matrix : 10-20% MA on typically MgO
  - At the periphery of the core **with low temperature**
    - » Retention of Helium
    - » Impact on Gaseous Swelling
    - » No large Modification of Oxygen Potential
    - » No large Modification of O, Pu, MA Redistribution
    - » Impact on thermal conductivity

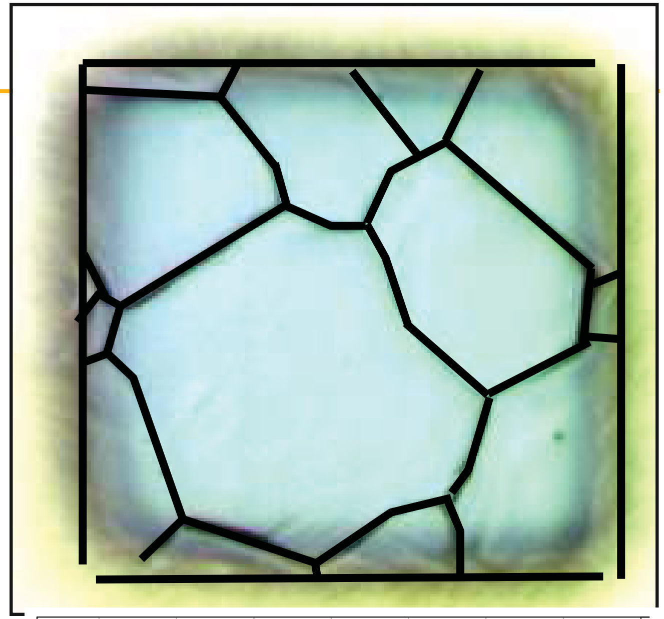
- Needs to model retention / release of helium and associated swelling
- Needs mechanical behavior of inert matrix
  - » Mechanics of heterogeneous microstructure
  - » He (and FP) implantation in inert matrix
- Very long irradiation time
  - » Impact on material properties of high damage

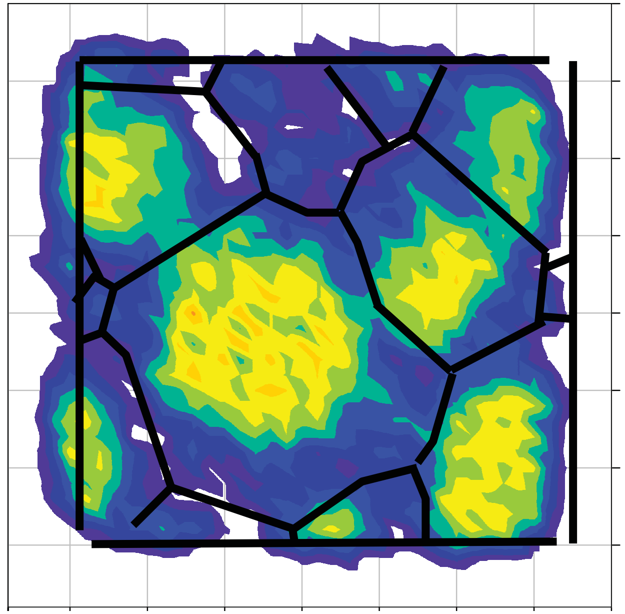
He retention in UO_2

On He implanted UO2 and NRA characterization

Labrim et al. NIMB 261 (2007)







At low dose

- Below 800°C : low He mobility
- Above 800°C :
  - Near grain boundary : defects annealing and large He mobility
  - In grain : bubble formation

At large dose

- Below 1000°C : low He mobility - bubble formation
- Above 1000°C : large He mobility

Needs for code improvement and validation

- **Models needs physical properties and behavior laws**
  - Analytical experiments
    - On simulated fuels to understand basic physico-chemicals behavior
  - Analytical irradiation
    - Some characteristics are fixed or adjusted during the irradiation and must be known also as function of radial (and axial) position.
  - Integral or semi-integral irradiation
    - Characteristics needed as function of radial and axial position.
    - in the worse case after the fabrication and during PIE and <u>in the best case all along the irradiation.</u>

FUEL MODELING VALIDATION: characteristics of interest

- **Irradiation conditions:**
  - Flux, spectrum, dose, power, temperature and thermal gradient, temperature and flow rate of the coolant
- **Composition :**
  - Fuel composition : wt% TRU, fission products, gases (helium and fission gases), impurities, stoechiometry (O/M)
  - Chemical form of each component : inside the fuel (single or multiple phases), interaction with others components (specially cladding), clad wastage
  - Gas composition and pressure in the gap
- **Microstructure:**
  - Density, porosity (open, closed, shape of porosity), grain size, columnar grain diameter, bubbles interconnection, cracks,
- **Geometry:**
  - Fissile column length, pellet diameter, inner/outer clad diameter, fuel central hole, gap closure
- **Properties (mainly but may be completed) :**
  - Thermal conductivity and thermal expansion, melting point, eutectics, Young modulus, creep.

Conclusion for modeling fuel with MA

- ***Improvement on modeling MUST BE COMPLETED with***
  - *More accuracy on fabrication data and irradiation conditions*
  - *Analytical experiments*
  - *More instrumentation in core, especially in MTR (also in prototype?)*
  - *More characterizations in hot cell*

      » ***To have good assessment of physical properties and behavior laws***

Atomistic modeling for innovative fuels

M. Freyss, B. Dorado, M. Bertolus, G. Martin
R. Skorek, S. Maillard, P. Garcia,

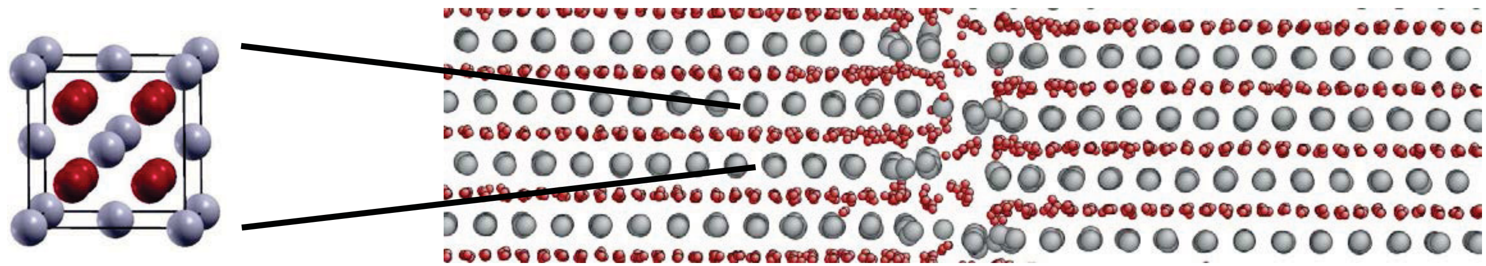
Commissariat à l'Energie Atomique
et aux Energies Alternatives (CEA)

Centre de ***Cadarache***
Fuel Study Department
Saint-Paul lez Durance, France

Context

- Increasing expectations for the modelling in order to improve existing nuclear fuels (**U-Pu oxides**,….) or to develop new innovative fuels (**U-Pu carbides**, **transmutation targets**…)

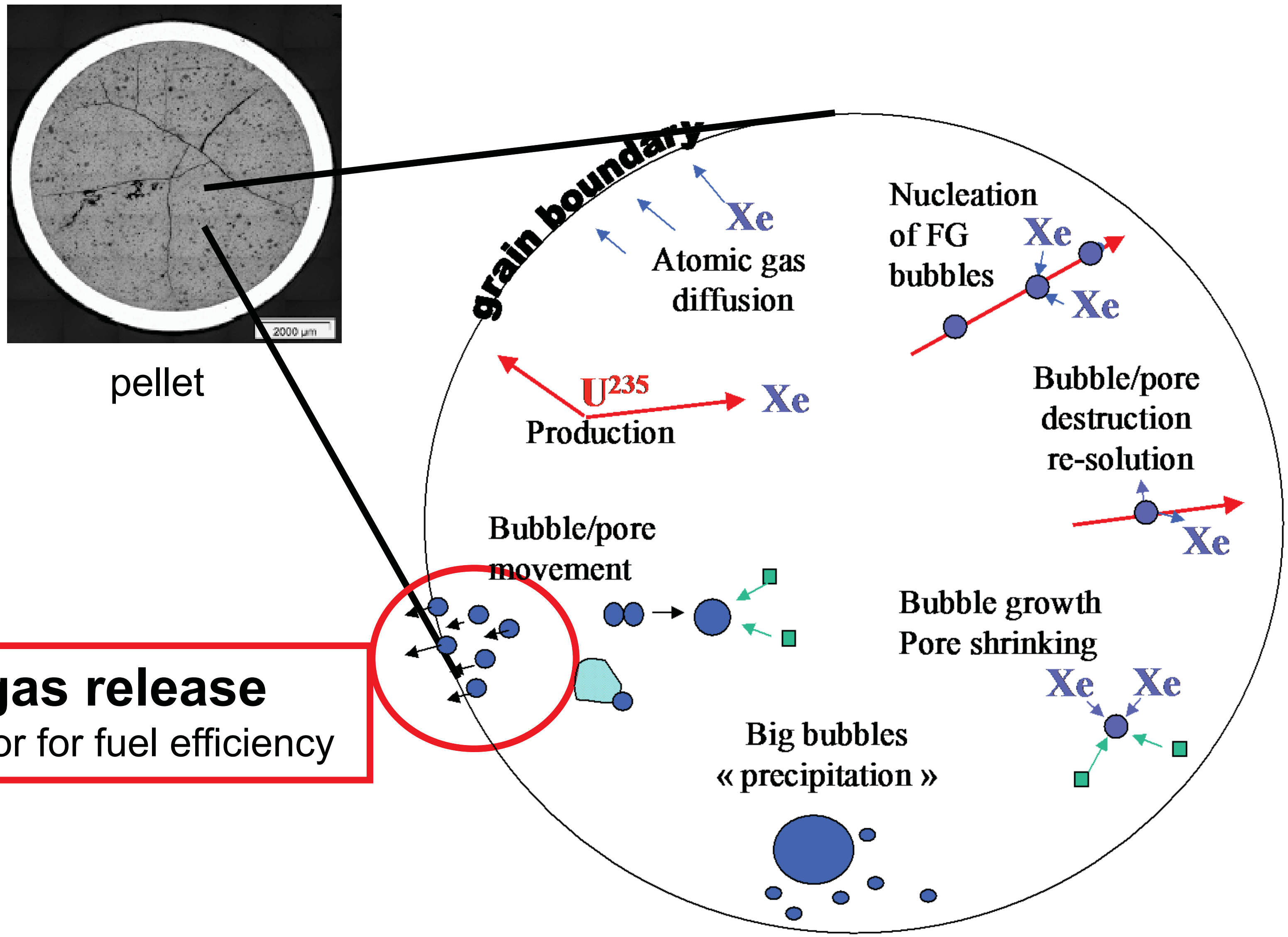
- Atomistic modeling to get insight into **elementary mechanisms** of the behavior of point defects, fission products… at the atomic scale



- Direct links between methods (**ab initio**, **MD**, **KMC**, **CD**…) to take more realistically into account the microstructure of the material (grain boundaries, fission gas clusters, ….). **Multi-scale approach** needed for the material modeling

- <u>**Reliable**</u> point defect formation and migration energies required

Fission gas diffusion in nuclear fuels



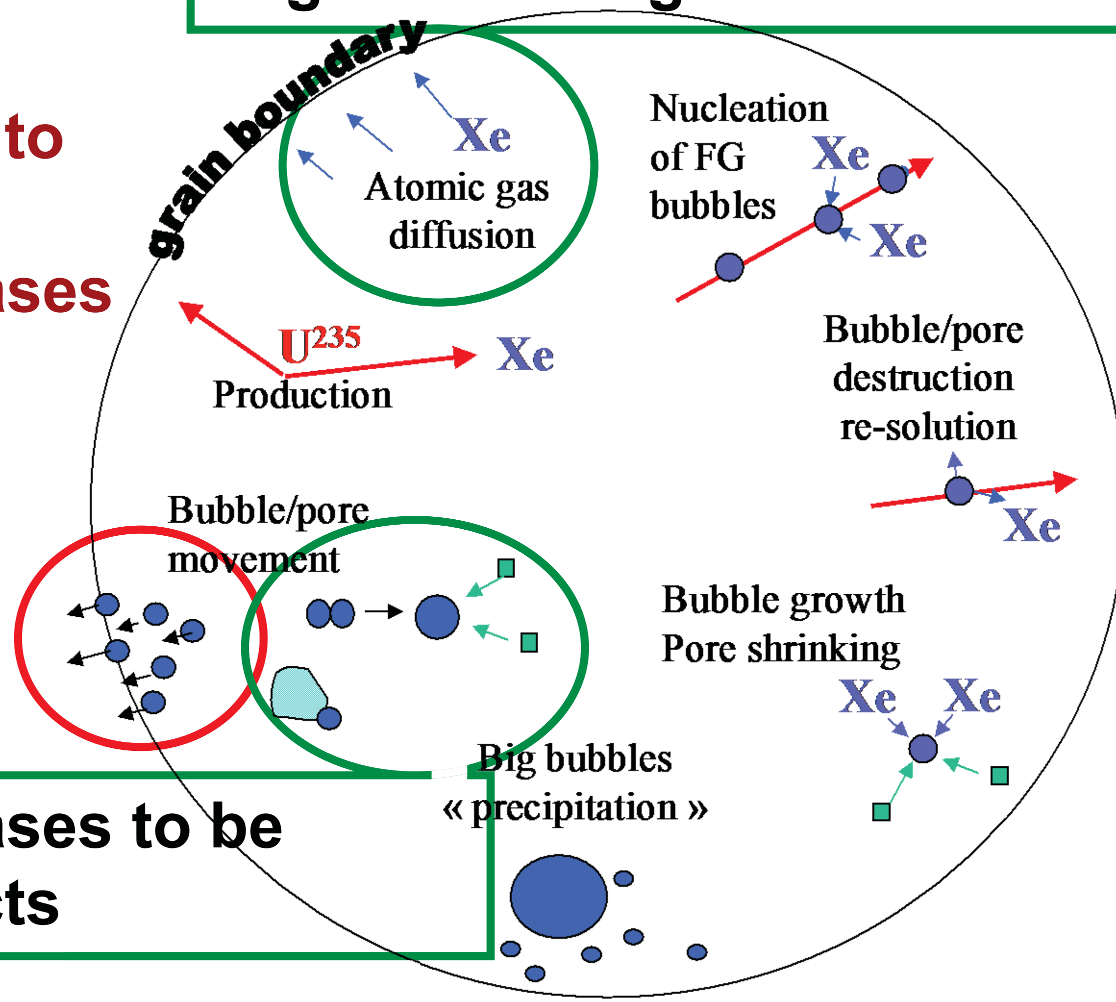
Fission gas release

Limiting factor for fuel efficiency

Fission gas diffusion in nuclear fuels

Tendency of fission gases to migrate in the grain

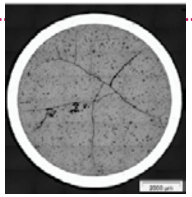
Atomistic calculations to get insight into the behaviour of fission gases at the atomic scale



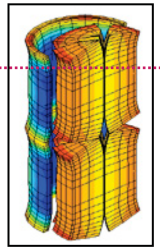
Tendency of fission gases to be trapped by point defects

Multi-scale modeling of nuclear fuels

Macroscopic scale



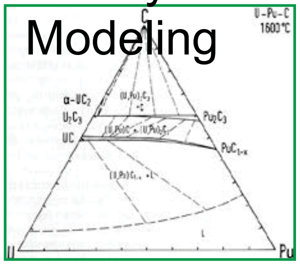
Pellet



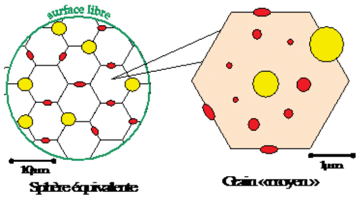
•Modeling of fuel behavior

Microscopic scale

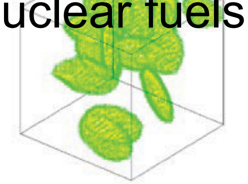
• Thermodynamics Modeling



• Modeling of fission gas behavior

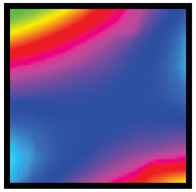


• Thermo-mechanical behavior laws of nuclear fuels

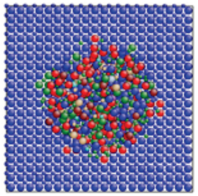


Atomic scale

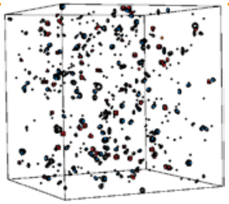
• Atomistic modeling of nuclear fuels:
structure, defect stability and fission gas behavior



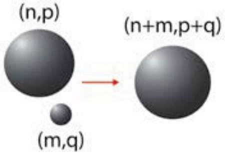
ab initio



DM



KMC

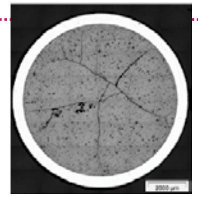


Cluster dynamics

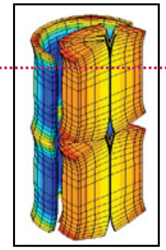
Coupling to experimental studies

Multi-scale modeling of nuclear fuels

Macroscopic scale



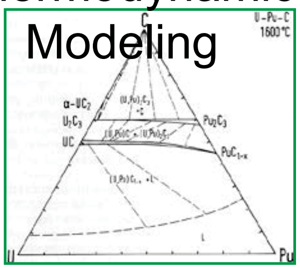
Pellet



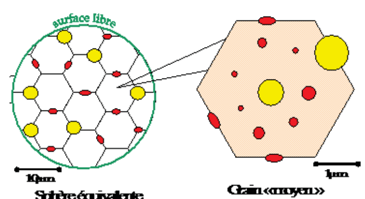
•Modeling of fuel behavior

Microscopic scale

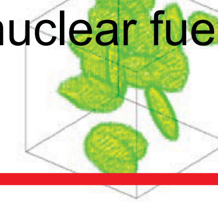
- Thermodynamics Modeling



- Modeling of fission gas behavior



- Thermo-mechanical behavior laws of nuclear fuels

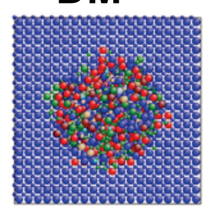


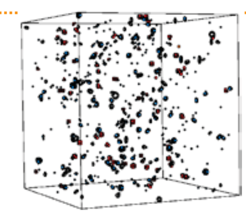
Atomic scale

- Atomistic modeling of nuclear fuels: structure, defect stability and fission gas behavior

ab initio

DM





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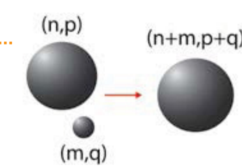


Cluster dynamics

Coupling to experimental studies

Ab initio modeling: Density Functional Theory

Quantum description of interaction between nuclei and electrons

Schrödinger equation

$$H\,\Psi(\vec{r}) = E\,\Psi(\vec{r})$$

Impossible to solve for systems with more than 1 electron **!**

Method to solve it: transform it into a single electron problem

Wave function Ψ for N electrons ➔ *Wave functions for 1 electron φ_i*

But retain description of the electronic interaction: important in bonding

$$E[n] = T_o[n] + V_H[n] + V_{ext}[n] + V_{xc}[n]$$

kinetic energy

electrons-electrons

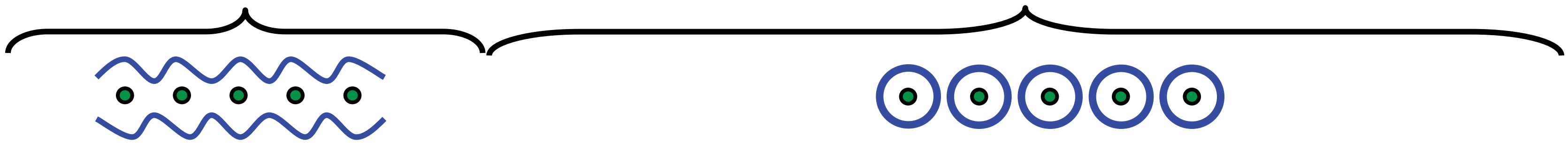
electrons-nuclei

exchange-correlation

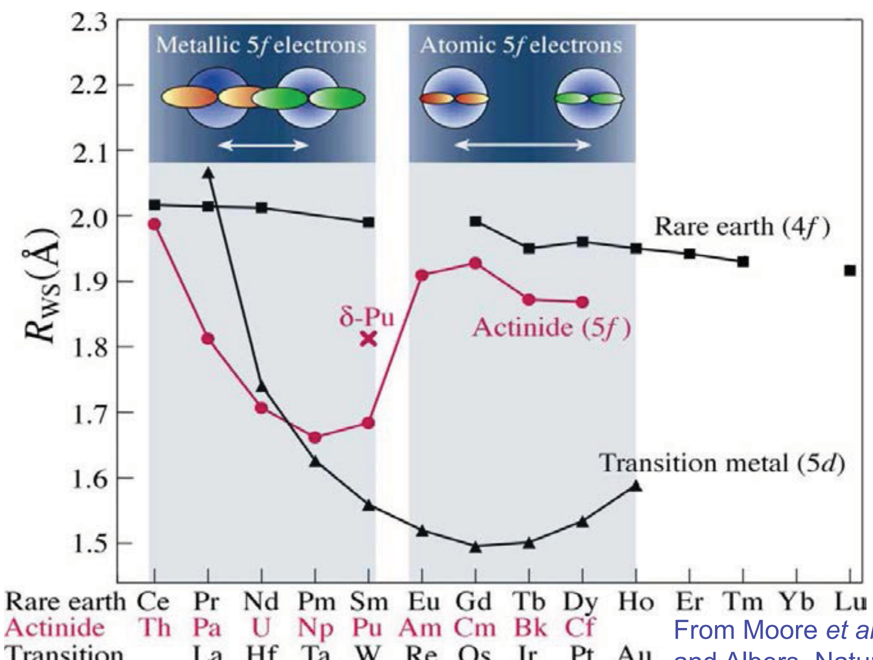
Ab initio modeling: Density Functional Theory

5f electron spatial localization in the actinide series

delocalized localized



90	91	92	93	94	95	96	97	98	99	100	101	102	103
Th	**Pa**	**U**	**Np**	**Pu**	**Am**	**Cm**	**Bk**	**Cf**	**Es**	**Fm**	**Md**	**No**	**Lr**



From Moore *et al.* Rev. Mod. Phys. 81, 235 (2009) and Albers, Nature **410**, 759 (2001).

UO_2, PuO_2, AmO_2 …:

insulator, **localized** *5f* electrons, strong correlations

underestimated by the DFT

Approximations beyond the standard DFT approximations

⇨ **DFT + U**

Dorado *et al*., Phys. Rev. B 79, 235125 (2009)

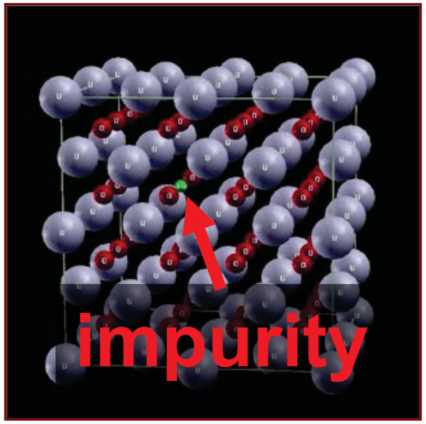
Point defects and fission products in AnO_2

- **Type of defects**

Vacancies, interstitials
Frenkel pairs *(1 int. + 1 vac.)*
Schottky defects
(1 vac. An + 2 vac. O)

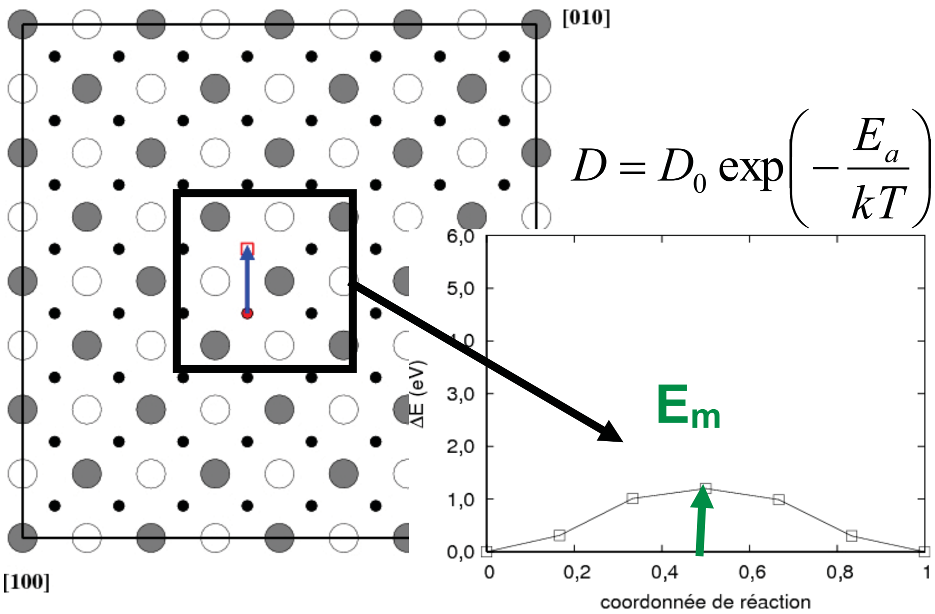
Fission gases and helium
kryton, xenon, iodine

2x2x2 cubic supercells



Stability: formation energies, incorporation energies
Migration: migration energies using the *nudge elastic band* (**NEB**) method

$$D = D_0 \exp\left(-\frac{E_a}{kT}\right)$$



UO_2: Dorado *et al.*, Phys. Rev. B 83, 035126 (2011)
UC: Freyss, Phys. Rev. B 81, 014101 (2010)

Extension to other An oxides:
PuO_2, AmO_2 Freyss *et al.* JNM (2006)
PuO_2,NpO_2 Tiwary *et al.* Phys. Rev. B **83**, 094104 (2011)

Empirical potentials for UO_2

Empirical potentials : **rigid ions model** (fixed point charges)

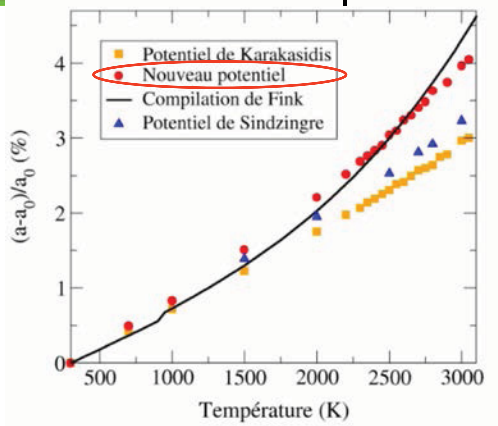
Buckingham interatomic pair potential:

$$V(r) = A\exp\left(-\frac{r}{B}\right) - \frac{C}{r^6}$$

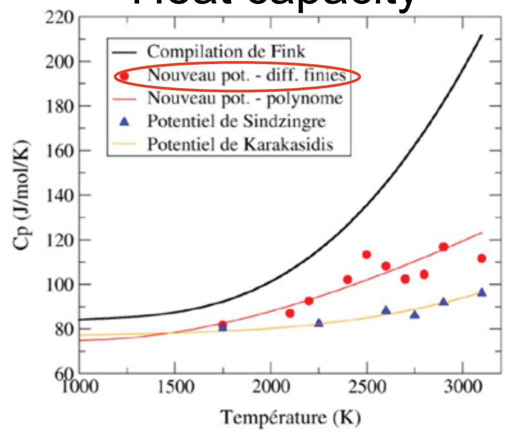
short-range repulsion

long-range dispersion

Thermal expansion



Heat capacity



Energy [eV]	Experimental	Ab initio GGA	Ab initio GGA+U	Rigid ions Morelon
Formation Frenkel O	3.5±0.5	3.3	5.3	3.9
Migration vac. O	0.5	1.2	0.7	0.3
Migration int. O (direct)	0.8 - 1.0	3.6	3.2	1.4
Migration int. O (indirect)		1.1	0.9	0.7
Formation Frenkel U	9.5	9.3	15.8	15.7
Migration vac. U	2.5	4.4	5.5	3.9
Migration int. U	2.0	5.8	/	4.2
Formation Schottky	6.5±0.5	4.1	2.5	3.9

GGA: M. Freyss *et al.* J. Nucl. Matter. 347, 44 (2005)
GGA+U: B. Dorado, PhD Thesis, CEA Cadarache (2010)

Parametrization (**A, B, C**) by **Morelon** (CEA):
bulk properties, *ab initio* formation energies of point defects, thermo-elastic data…

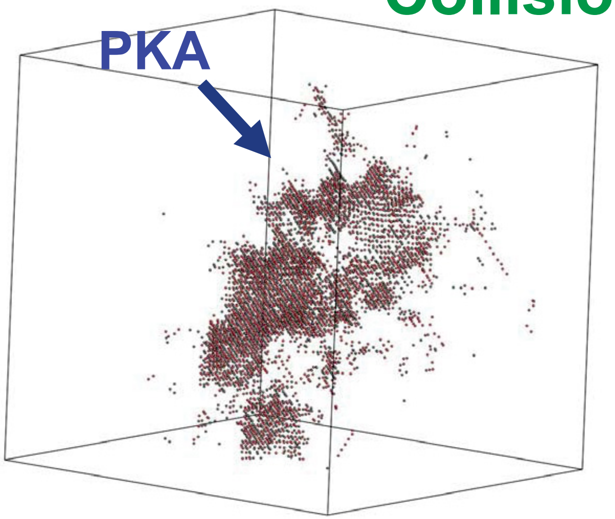
Morelon *et al.*, Phil. Mag. 83, 1533 (2003)

$(U,Pu,Np)O_2$ Tiwary *et al.* Phys. Rev. B **83**, 094104 (2011)

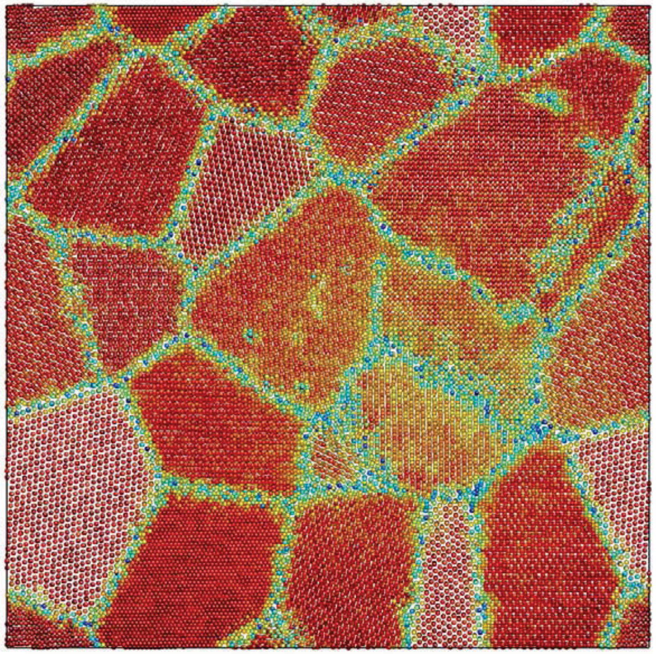
AmO_2 Uchida *et al.* JNM **400**, 3 (2010)

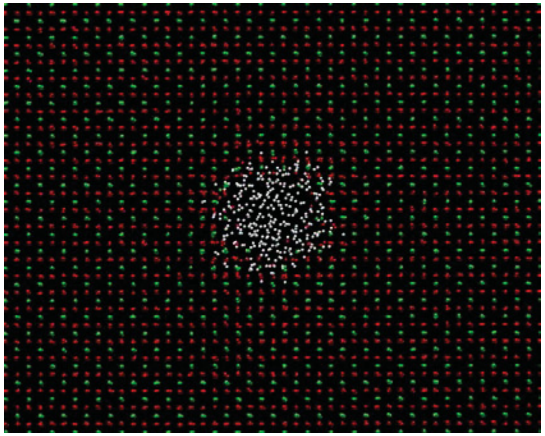
Empirical potential modeling

Collision cascades



Defects at grain boundaries

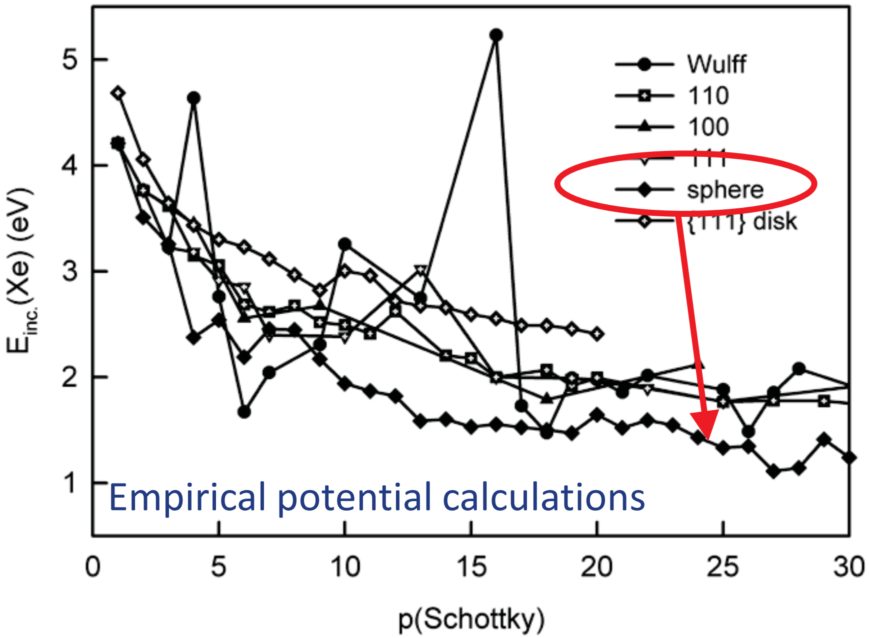




Gas bubble nucleation

Radiation damage and stability of Xe clusters in UO_2

Xenon incorporation energies in different void shapes, as a function of the number *p* of Schottky defects, which contain *p* Xe atoms

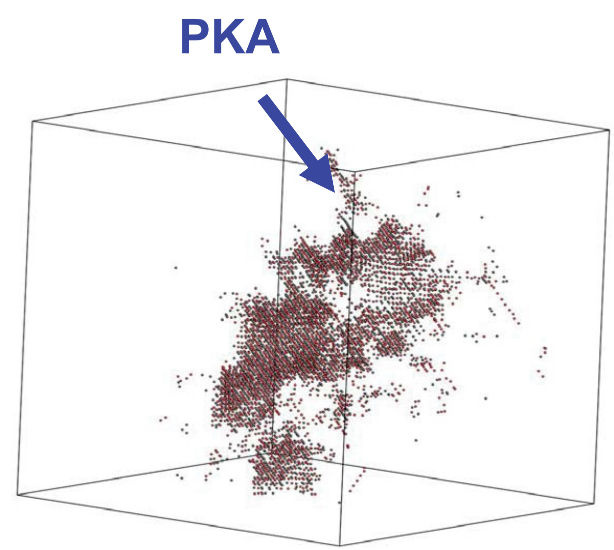


→ Spherical xenon clusters with **radius of ~ 1.3 nm**. In agreement with TEM and SAX analysis

A. Chartier, L. van Brutzel. M. Freyss, Phys. Rev. B 81, 174111 (2010)

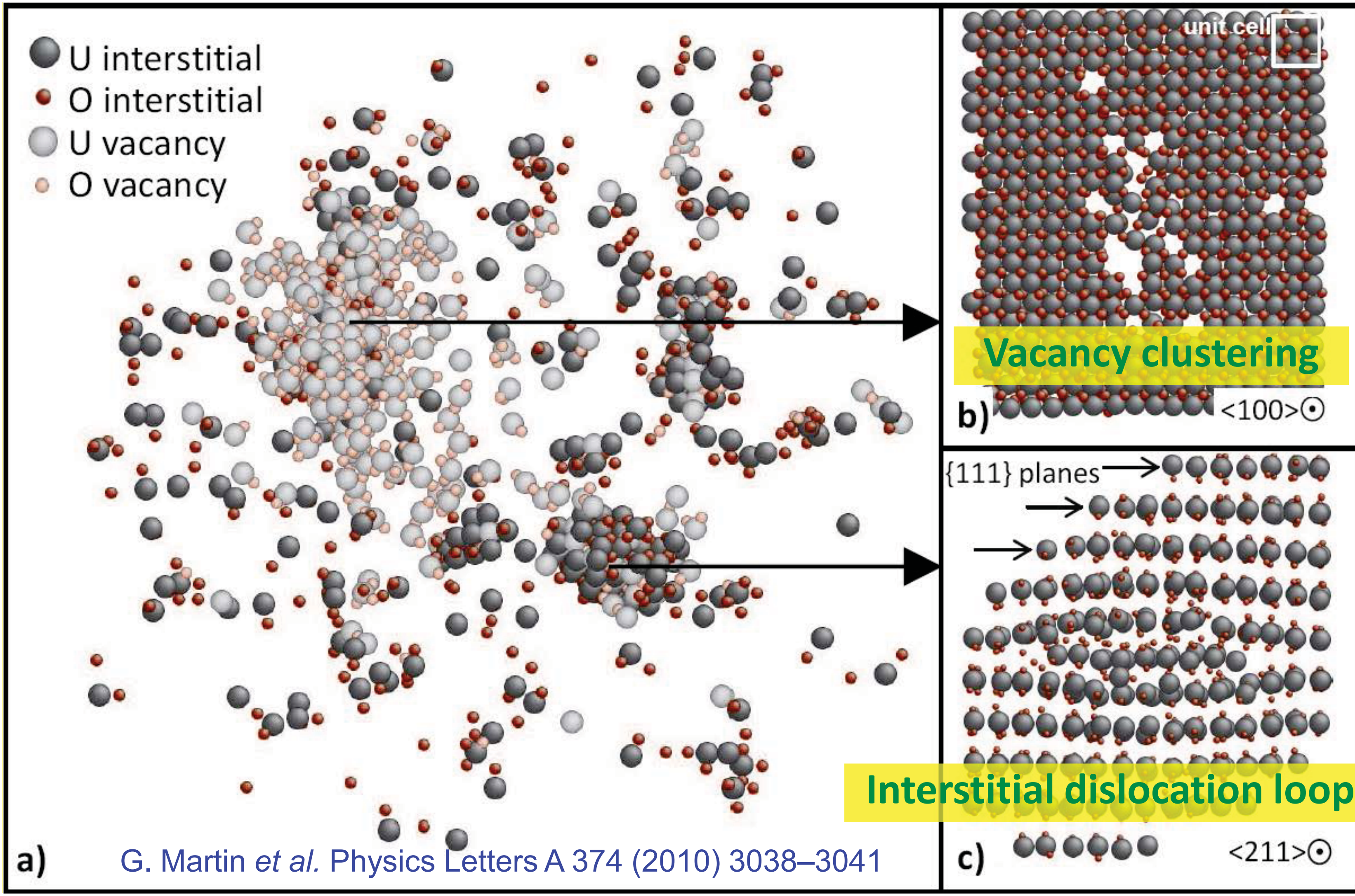
Damage formation after displacement cascades: energy pulse given to an atom (Primary Knock-on Atom PKA) → 1 to 80 keV

Ballistic damage created by the slowing-down of recoil nuclei



G. Martin *et al.*, Journal of Nuclear Materials **385**, 351 (2009)

Damage production after cascades in UO_2



Coupled with MET analysis of ion implanted UO_2 samples (thesis of Amélie Michel)

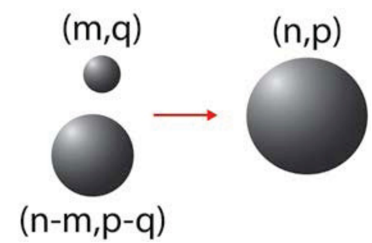
Cluster dynamics

Several models exist to model **FG behaviour** → e.g. MARGARET at CEA

Most observation are correctly simulated but some phenomena are **empirically** modelled (not based on a mechanistic comprehension), in particular **bubble sink strength** is empirically changed for annealing simulations

⇨ Cluster Dynamics (= rate theory model)

Comprehensive framework to calculate defect and defect clusters (bubbles / loops) **concentrations** over time, with time scale and length scale appropriate for fuel study *(years, grains)*



Schottky (n < 0)
Anti-Schottky (n > 0)

n = number of self-defects

p = number of solutes

Xe

$$\frac{\partial C(n,p,t)}{\partial t} = \boxed{\sum_{n',p'} J_{(n',p')\rightarrow(n,p)}} - \boxed{\sum_{n',p'} J_{(n,p)\rightarrow(n',p')}} + \boxed{G(n,p)} - \boxed{L(n,p)}$$

Transition rate of reactions toward (n,p)

Transition rate of reactions from (n,p)

Source term

Sink term

Input parameters : *D, E_b, E_m,…*

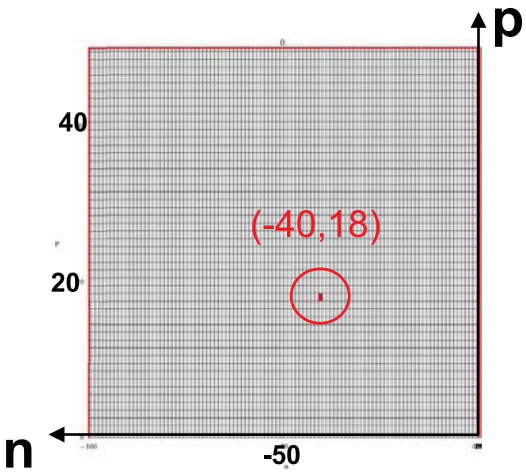
Cluster dynamics: gas bubble growth

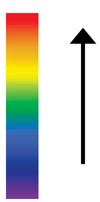
Schottky migration energy	2.5 eV	
Xe - Schottky migration energy	2.5 eV	
Schottky formation energy	2.5 eV	
Xe incorporation energy within a Schottky	8.0 eV	1.8 eV

Annealing time	10 min
Temperature	1400°C

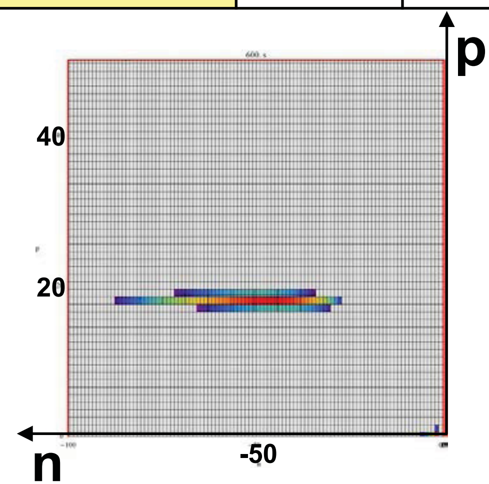
Initial conditions

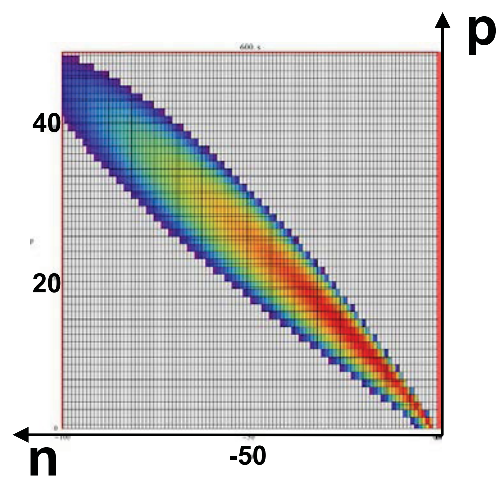
C(-40,18) = 10^{17}cm^{-3}
No source term and no sink term
Mobile clusters: (-1,0) and (-1,1)



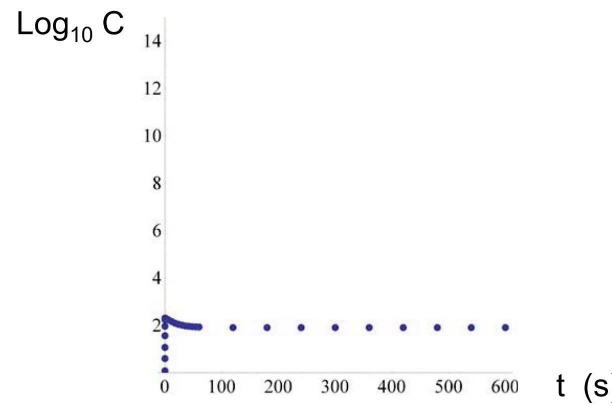


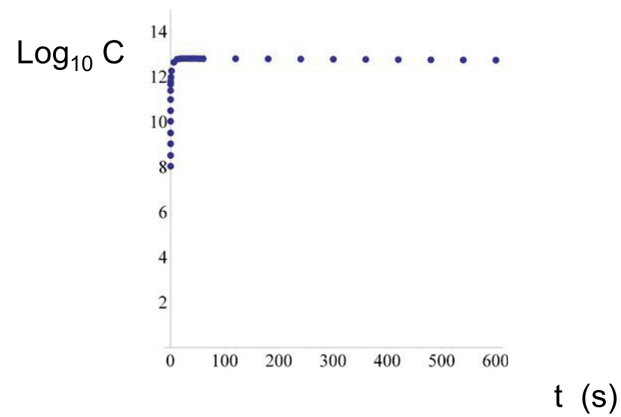
Increasing bubble concentration





C(-1,1) over time





Need of experiments for validation

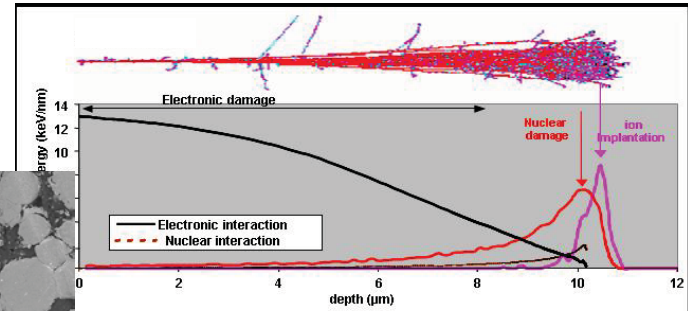
Each modeling technique requires experimental validation:
- for the **approximations** or **models** used
- for the physical data used as **input material properties**

Not much data available for mixed oxide fuels such as $(U,Pu,MA)O_2$ and for the fission gas behavior in those materials

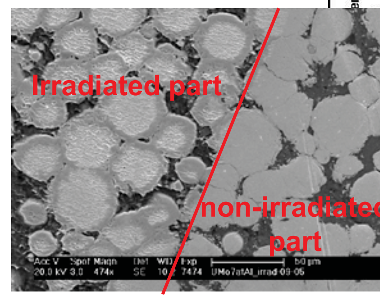
Modeling approaches	Observations	techniques
Ab initio	- location of fission products - activation energies of diffusion (self-diffusion and gases)	- TEM, XAS,... - SIMS, ...
Empirical potentials	- material thermo-mechanical properties - radiation damage, microstructure evolution - fission gas and helium bubbles	- SEM-FIB, MET, PAS, EBSD,... - SAX, TEM,...
Cluster dynamic	- atomic diffusion coefficients - size and density of fission gas and helium bubbles, …	- TDS, SIMS,... - SAX, TEM,...

Seperate effect studies of nuclear fuels

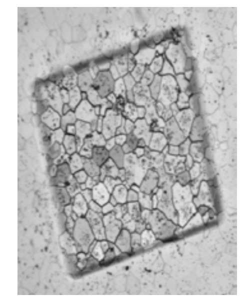
- Non active model materials such as depleted UO_2
- Ion **implantation** to simulate fission products

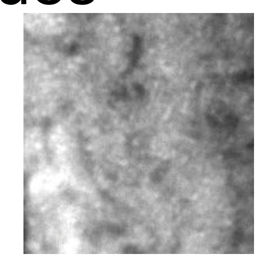


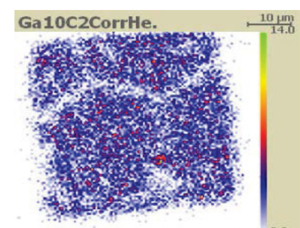
- **Thermal treatment** or **heavy ion irradiation**



- **Characterization** with a large panel of dedicated techniques (SIMS, RBS, NRA, TEM, XAS)







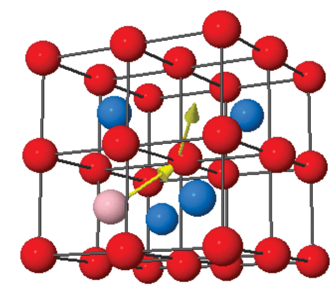
- **Large scientific facilities** (particle accelerators and synchrotron radiation)

Oxygen migration: comparison to experimental results

Comparison to experimental results: **Electrical conductivity measurements + SIMS experiments** + control of parameters that affect the material (oxygen partial pressure and impurity content):

➢ **Oxygen diffusion occurs via interstitial mechanism**
Garcia *et al.*, J. Nucl. Mater. 400, 112 (2010)

➢**DFT+U calculations** show that **oxygen diffusion in UO_2 occurs *via* an interstitialcy mechanism** when O diffusion is governed by interstitials


Jmol

Experimental value of the diffusion activation energy E_a:

$$\frac{D}{\sqrt{p_{O_2}}} \propto \exp\left(-\frac{E_a}{kT}\right) = \exp\left(-\frac{E_F + E_m}{kT}\right)$$

Experimental value E_a **= 0.75 ± 0.08 eV**
Calculated migration energy: E_m = 0.93 eV
Calculated formation energy: E^F = - 0.05 eV
Calculated activation energy: $E_a = E^F + E_m$ = **0.88 eV**

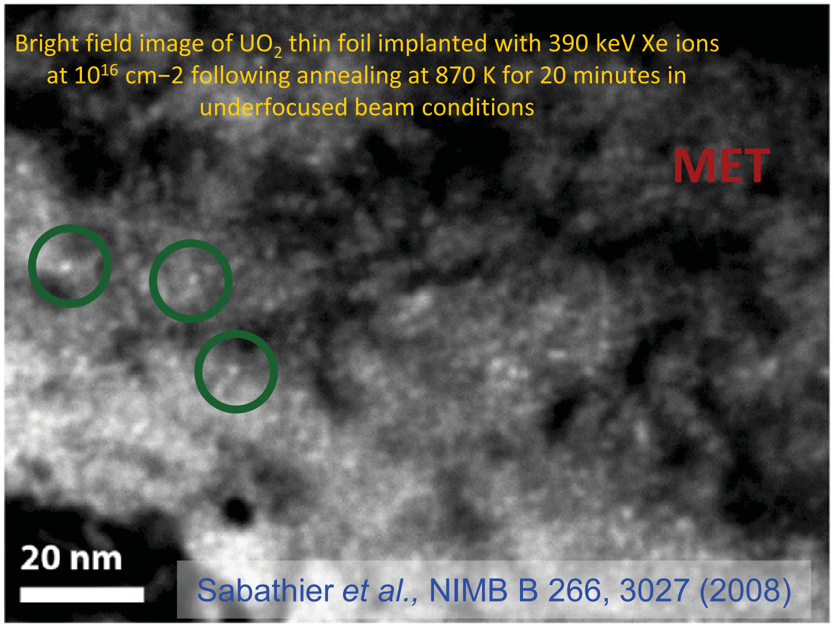
Dorado et al., Phys. Rev. B 83, 035126 (2011)

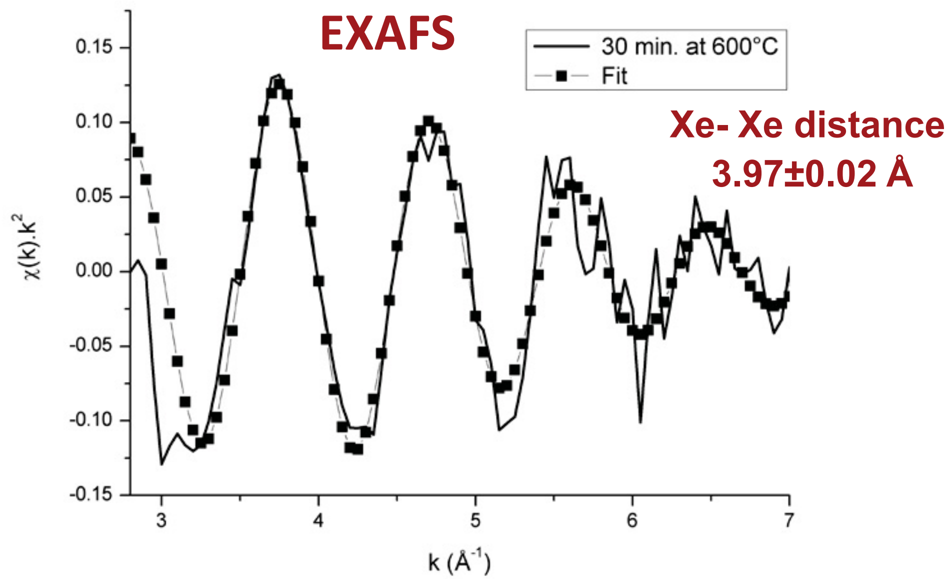
Good agreement between experimental and calculated values

XAS and TEM characterization of Xe bubbles in UO_2

Ph. Martin
C. Sabathier

- 2 at .% Xe implantation in UO_2 polycrystalline samples
- annealing 20-30 minutes at **600°C**





Xe aggregate size ~ 2 nm

d_{Rg-Rg} → **lattice parameter** →(E.O.S.) **aggregate internal pressure**

P=f(V,T) by K. Asaumi, Phys Rev. B 29(1984))

P ~ 2.8 ± 0.3 GPa

In agreement with empirical potentials calculations

Chartier, van Brutzel, Freyss, Phys. Rev. B 81, 174111 (2010)

P. Garcia *et al.*, J. Nucl. Mater. **352**, 136 (2006)
P. Martin *et al* NIMB. B **266**, 2887 (2008)

Conclusions

- Modeling of the standard UO_2 fuel behavior much improved
- Atomistic modeling of fuels has much benefited from the improvement of the methods/approximations and of the increasing computing power
- Already very **favourable** and **encouraging confrontation of atomistic calculations** and **experimental results for UO_2** (oxygen migration, xenon bubble nucleation, …)
- Data transfer between modeling techniques
- Similar modeling for **(U, Pu, MA)O_2**, **(U, MA)O_2**, **(MA)O_2+IM** and carbides
- **Quality and accuracy of the calculated results can only be ensured provided some experimental data are available**

Acknowledgments

CEA: B. **Amadon,** G. **Jomard,** M. **Torrent,** F. **Jollet,** L. **van Brutzel,** A. **Chartier,** A. **Barbu**

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Los Alamos National Lab: D. **Andersson**

Los Alamos NATIONAL LABORATORY EST. 1943

F-BRIDGE project

F-BRIDGE

Basic **R**esearch for **I**nnovative **F**uel **D**esign for **GE**N IV systems

Computing facilities:

GENCI

Grand Equipement National de Calcul Intensif

GENCI GRAND EQUIPEMENT NATIONAL DE CALCUL INTENSIF

CCRT

Centre de Calcul Recherche et Technologie

ccrt

PIE Needs to Support Fuel Design & Safety Testing

Steven L. Hayes, PhD

Nuclear Fuels & Materials Division
Idaho National Laboratory

Outline of Presentation

- **Fuel Development & Safety Testing**
  - Historic process
  - US (DOE) development and qualification of a new fuel
- **Postirradiation Examination Needs**
  - Traditional suite of exams (largely macroscopic)
  - Non-traditional microscopic characterization
  - Emerging need for nano-scale characterization
- **Conclusion**

FUEL DEVELOPMENT
&
SAFETY TESTING

Fuel Development & Safety Testing

Crawford, et al., **Journal of Nuclear Materials**, **371**: 232-242 (2007).

- **Review of 4 phases of fuel development process**
  1) Fuel candidate selection
  2) Concept definition and feasibility
  3) Design improvement and evaluation
  4) Fuel qualification and demonstration
- **Process has decades of experience for EBR-II & FFTF fuel qualification/confirmation testing**

TRL Objective in Fuel Qualification

TRL	Function	Definition
1		A new concept is proposed. Technical options for the concept are identified and relevant literature data reviewed. Criteria developed.
2		Technical options are ranked. Performance range and fabrication process parametric ranges defined based on analyses.
3		Concepts are verified through laboratory-scale experiments and characterization. Fabrication process verified using surrogates.
4		Fabrication of samples using stockpile materials at bench-scale (~100 gram batches). Irradiation testing of small-samples (rodlets) in relevant environment. Design parameters and features establsihed. Basic properties compiled.
5		Fabrication of pins using prototypic feedstock materials at laboratory-scale (10 kg). Pin-scale irradiation testing at relevant environment. Primary performance parameters with representative compositions under normal operating conditions quantified.
6		Fabrication of pins using prototypic feedstock materials at laboratory-scale (1 kg) and using prototypic fabrication process. Pin-scale irradiation testing at relevant and prototypic environment (steady-state and transient testing). Safety basis establis
7		Fabrication of test assemblies using prototypic feedstock materials at engineering-scale (100 kg) and using prototypic fabrication process. Assembly-scale irradiation testing at prototypic environment. Safety basis established for full-core operations.
8	–	Fabrication of a few core-loads of fuel (tons) and operation of a prototype reactor with such fuel.
9		Routine commercial-scale operations. Multiple reactors operating

Fuel Development Phase 1: Fuel Candidate Selection

- **Objective: Based on previous experience, identify candidate fuel forms/concepts that appear capable of meeting mission needs.**
- **Selection criteria might include:**
  - Ability to accommodate desired fuel compositions
  - Experience with similar fuel types or analogues
  - Suitability of established fabrication techniques, or the potential for successful innovative techniques
  - Anticipated performance capabilities (e.g., temperature, burnup, or fluence)
  - Anticipated safety-related behavior (which may be quite speculative at an early stage)
  - Suitability of design, considering issues such as fuel-cladding compatibility, fuel-coolant compatibility, and fuel properties
  - Compatibility with envisioned back-end fuel cycle technology
  - Expected cost of fabrication
- **Achieves TRL 2**

Fuel Development Phase 2: Concept Definition and Feasibility

- **Objective: Establish a reference concept/design**
- **Determine how the fuel can be fabricated**
  - Process scoping & feasibility
  - Fabricate characterization and test samples
- **Determine and assess key properties**
  - Feasibility issues for irradiation testing
- **Use scoping irradiation tests to assess phenomena envisioned to impact feasibility and fuel lifetime**
  - Simple experiments
  - Prototypic conditions as much as possible
- **Achieves TRL 4**

Fuel Development Phase 3: Design Improvement and Evaluation

- **Objectives**:
  - Optimize the fuel design for economics, performance and safety
  - Produce a Fuel Specification and a Fuel Safety Case for a core of fuel
  - Establish predictive fuel performance code (or codes)
- **Develop engineering-scale fabrication processes and equipment**
- **Assess fuel properties vs. composition or processing variations**
- **Irradiation testing to:**
  - Determine sensitivity of performance to fuel design and fabrication variables and to operating conditions
  - Establish burnup limits and safety margins for various operating conditions (normal and off-normal)
- **Develop fuel behavior models and predictive codes**
- **Achieves TRL 6**

Fuel Development Phase 4: Fuel Qualification and Demonstration

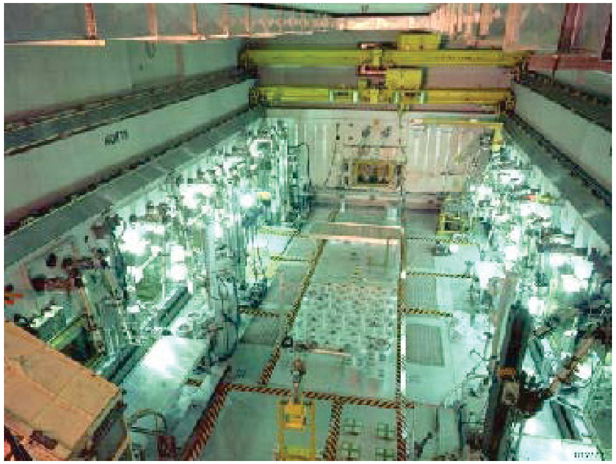
- **Objectives**:
  - Qualify production-line fuel as the driver fuel for a demonstration reactor
  - Demonstrate the safety and reliability of a core of fuel through successful operation of the demonstration reactor
  - Validate predictive fuel performance code (or codes)
- **Demonstrate production of fuel in conformance with Fuel Specification**
- **Demonstrate through LTA irradiation that fuel/fuel assembly behavior is within the bounds of the Fuel Safety Case**
- **Accumulate reactor performance data and operating experience with a core of fuel to support licensing of first-of-a-kind unit**
- **Achieves TRL 7 or 8**

POSTIRRADIATION EXAMINATION NEEDS

Traditional Suite of (PIE) Examinations

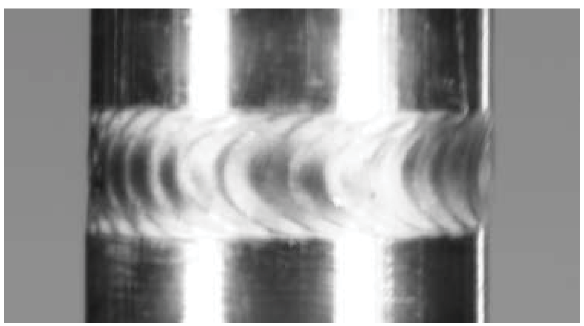
1) Non-Destructive Examinations

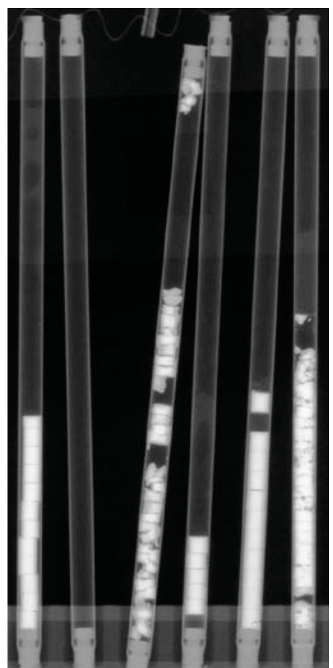
2) Destructive Examinations

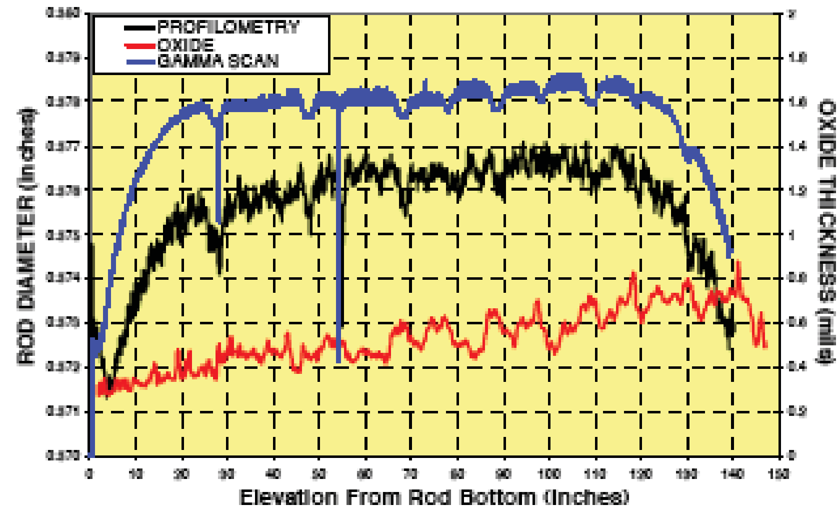


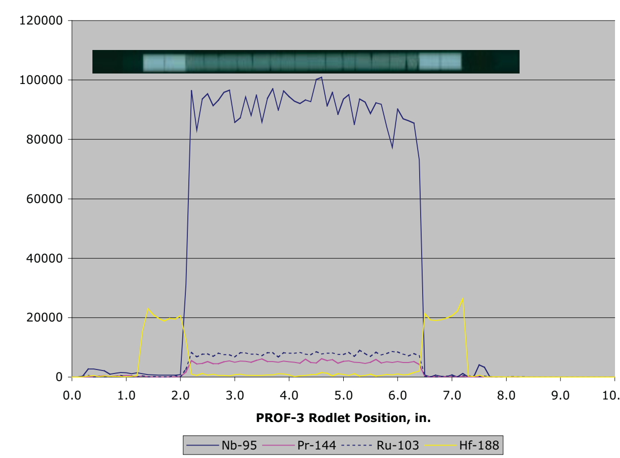
Non-Destructive Examinations

- ✓ Visual Inspection
- ✓ Neutron Radiography
- ✓ Dimensional Inspection
- ✓ Gamma Ray Spectroscopy
- ✓ Eddy Current Oxide Layer Tester
- ✓ Eddy Current Cladding Integrity Tester



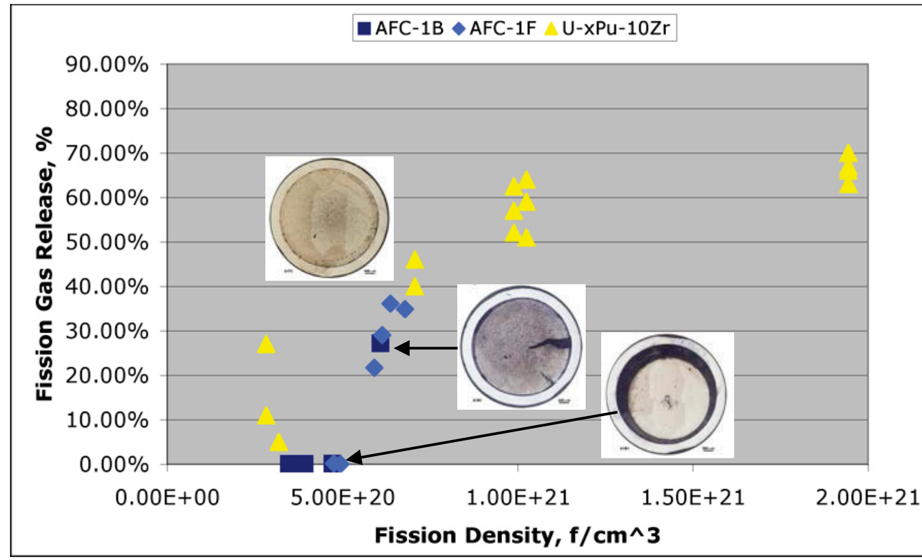


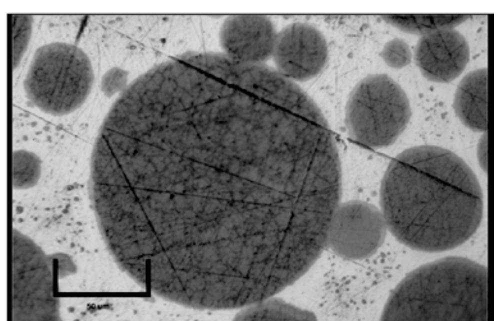


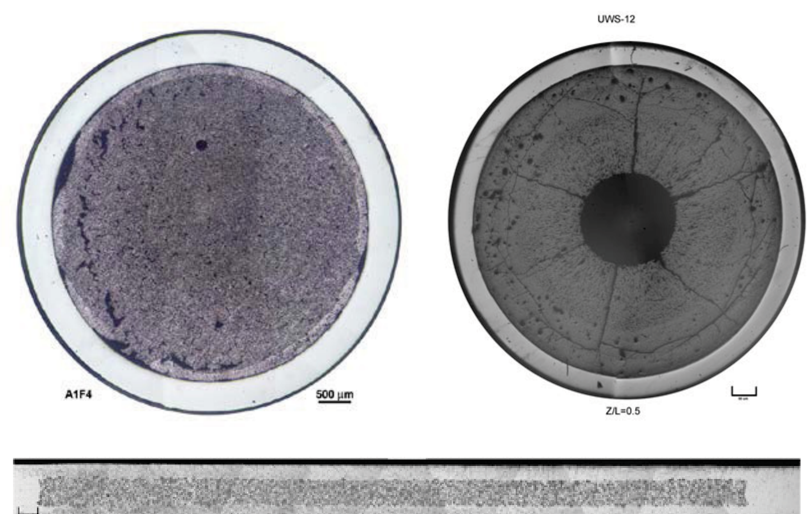


Destructive Examinations

- ✓ Plenum Puncture & Gas Analysis
- ✓ Isotopic & Burnup Analysis
- ✓ Metallography/Ceramography
- ✓ Physical Properties
  - *Fuel Density*
  - *Cladding Hydrogen Analysis*
  - *Cladding Mechanical Properties*
- ✓ Support for Safety Testing
  - *Handle/disassemble loops?*
  - *Fuel Annealing Furnace*

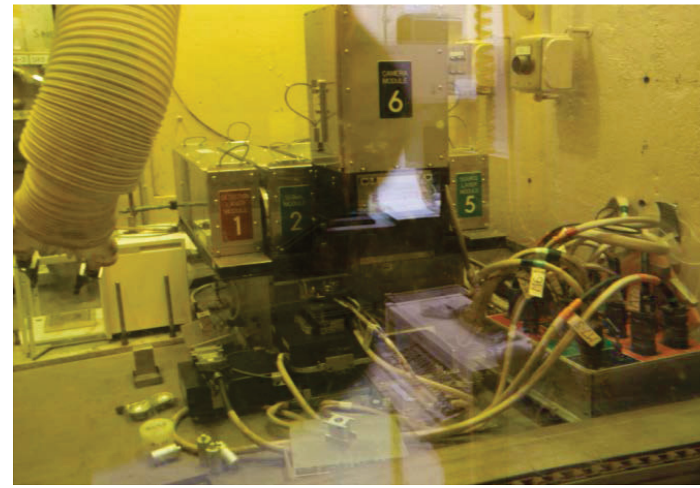


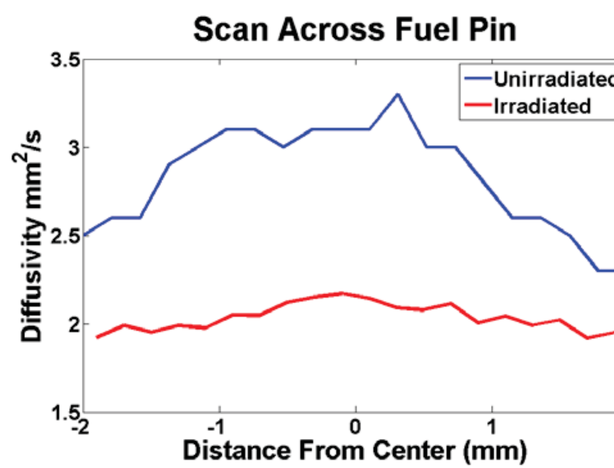




Non-Traditional Microscopic Characterization

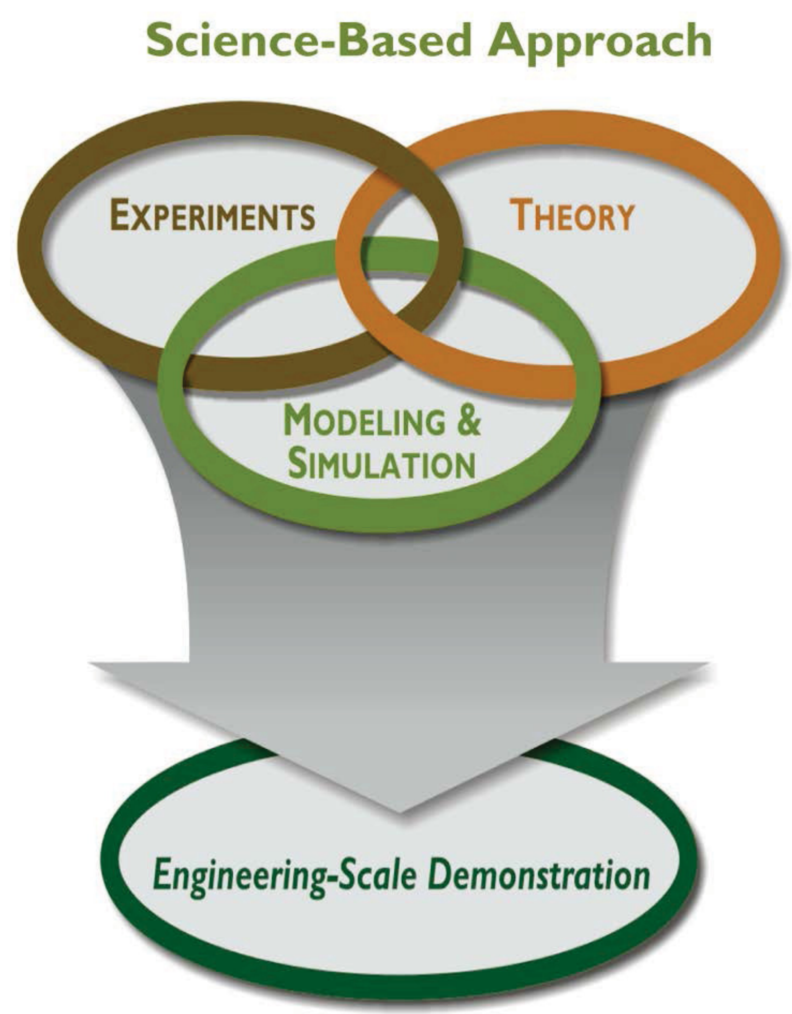
- ✓ Scanning Electron Microscopy
- ✓ Transmission Electron Microscopy
- ✓ Electron Micro-Probe Analysis
- ✓ Physical Properties
  - *Thermal Conductivity/Diffusivity*
  - *Melting Point*





Emerging Need for Nano-scale Characterization

- **Why needed/desired?**
  - Prototypic test environments no longer available (i.e., fast reactors, transient reactors)
  - Historic, empirically-based approach costly and time-consuming
- **Characteristics of approach**
  - Develop/apply theory to fuel behavior
  - Perform separate effects testing
    - *Guided, informed by theory*
    - *Extensive pre-irradiation characterization*
    - *Simulated with advanced M&S*
  - Detailed postirradiation examinations
    - *Inform models, validate theory*
- **Potential results**
  - Faster, less costly fuel development
  - Number of experiments reduced, but more effective



Conclusion

- **Historic process to develop/qualify new fuels is proven**
  - Costly and time-consuming process
  - Necessary infrastructure (in some cases) no longer exists

- **Future development of new fuels will likely evolve**
  - Historic (empirically-based) process will continue to be used
  - Supplemented with increasing dependence on tools developed under advanced modeling and simulation efforts

- **Conclusion**
  - Traditional PIE processes will still be needed
  - Development of micro- and nano-scale methods of characterizing irradiated nuclear fuels and materials will be increasingly important

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Wir schaffen Wissen – heute für morgen

Paul Scherrer Institut

Manuel A. POUCHON

Beam-line techniques

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- Introduction
  - Aspects of beamlines / Types of beamlines
- X-Ray beamlines
  - Synchrotron light sources
    - Basics / Beamlines in the world
    - Selection of analysis techniques
    - Examples
      - PIE of MOX by µXRD, EXAFS
      - XRD of ODS after He irradiation
  - Free electron laser (FEL)
    - Special aspects/Possible application
- Neutrons
  - Sources / Features
  - One example

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Aspects of beamlines

Advantages, special features:

- **High flux** (e.g. Synchrotron light, FEL)
- **Special particles** for analysis (e.g. neutrons, electrons, ions, muons ,...)
- Large **range of energies** (up to very high ones)
- Special features as
  - **polarization**
  - **coherency** (e.g. for X-Ray from FEL),
  - **pulsed sources** (e.g. FEL, neutrons, …)
  - high beam quality for superior **focalization** (for μ analysis) → option of **scanning**

→Evolving of **new experimental techniques** exploring more material features, physical/chemical effects

Disadvantages:

- Experiment requires **long preparation** time (with application phase) for a couple of hours up to a few days of experimental time (**low flexibility**)
- Beamlines **often** do **not accept radioactive samples**
- if **proprietary** research, then **very expensive**

Outline

Types of beam-lines

- X-Ray
  - Synchrotron light sources ←
  - Free electron laser ←
- Neutron
  - Reactors
  - Spalation ←
- Muons (Muon spectroscopy)
- Electrons (TEM, …)
- Ions (RBS, ERDA, …..)
-
-

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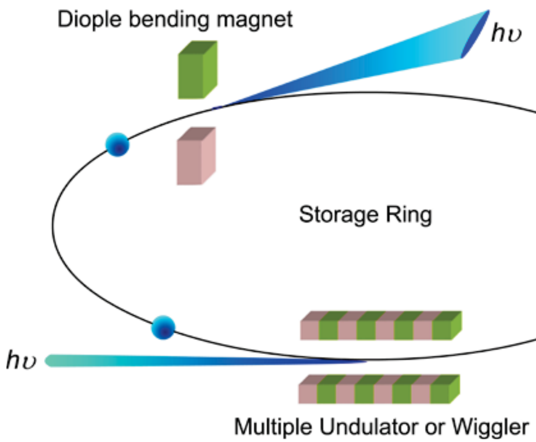
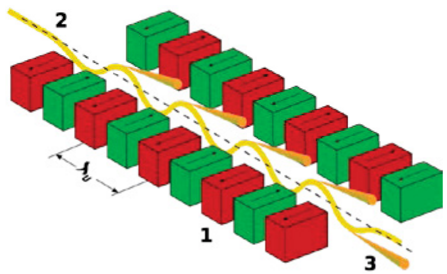
Synchrotron light sources





Synchrotron radiation

soleil , France

Synchrotron
- Intense light
- Infrared – Hard X-Ray (huge range)

http://en.wikipedia.org/wiki/Synchrotron_light

Synchrotron radiation experiments

Beamlines for radioactive samples

- ANL/APS – (USA)
- Berkeley Lab/ALS- (USA)
- BNL/NSLS – (USA)
- ESRF – (France) – element specific
- KIT/ANKA - INE – (Germany) – element specific
  C-Lab
- SLS – Microxas - (Switzerland) – 100 LA
- Soleil – Mars - (France) - <1 LA
- Spring8/JAEA - Actinide Science – (Japan)
- Stanford University/SSRL – (USA)

Sample downsizing

Local shielding

http://en.wikipedia.org/wiki/List_of_synchrotron_radiation_facilities

X-Ray techniques

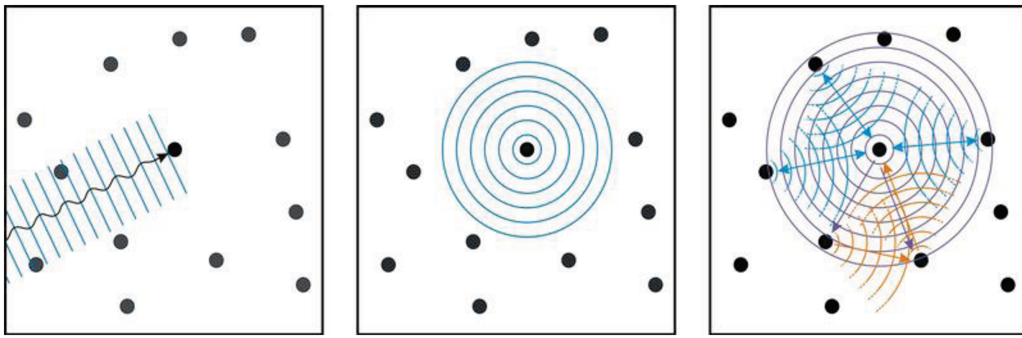
Possible techniques

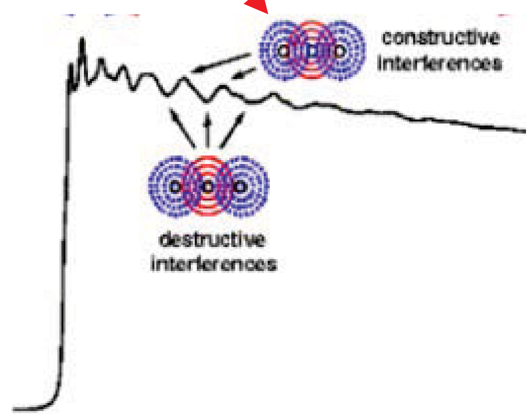
- X ray absorption techniques
  - EXAFS / XANES ←
- μ-XRD ←
- Tomography (high(er) resolution, penetration)
- Fluorescence
-
-
-

Explanation for X-Ray absorption techniques

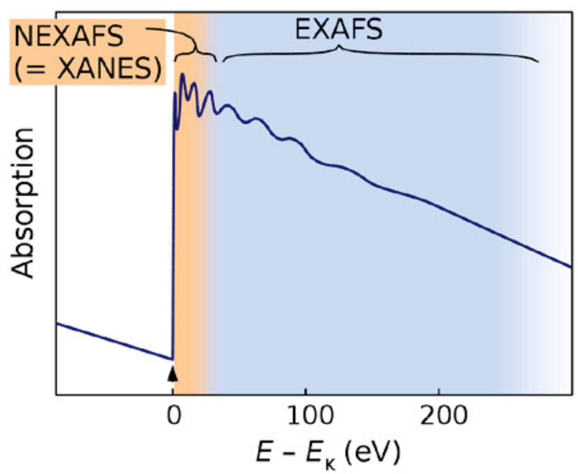
XANES/EXAFS

Absorption of incoming beam

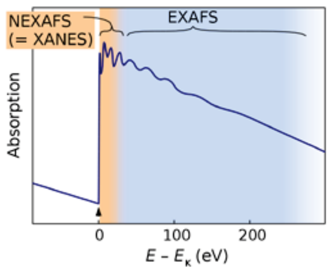




Resulting absorption spectrum



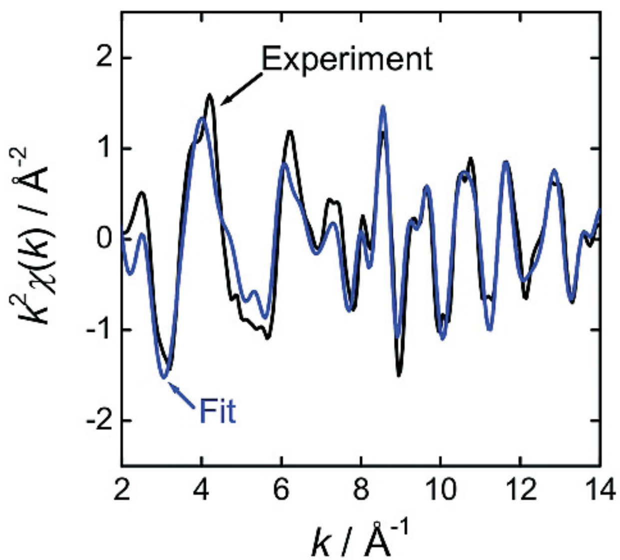
Explanation for X-Ray absorption techniques

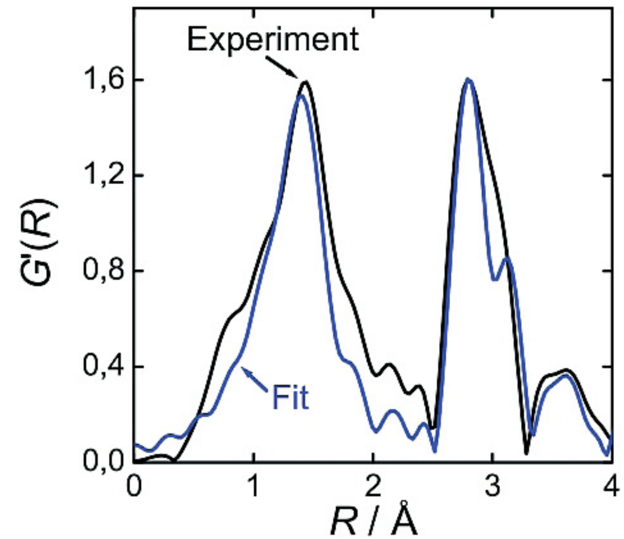


Extraction of **fine structrue** (substraction of atomic absoption)

Fourier trsf.

Radial distribution function (RDF)





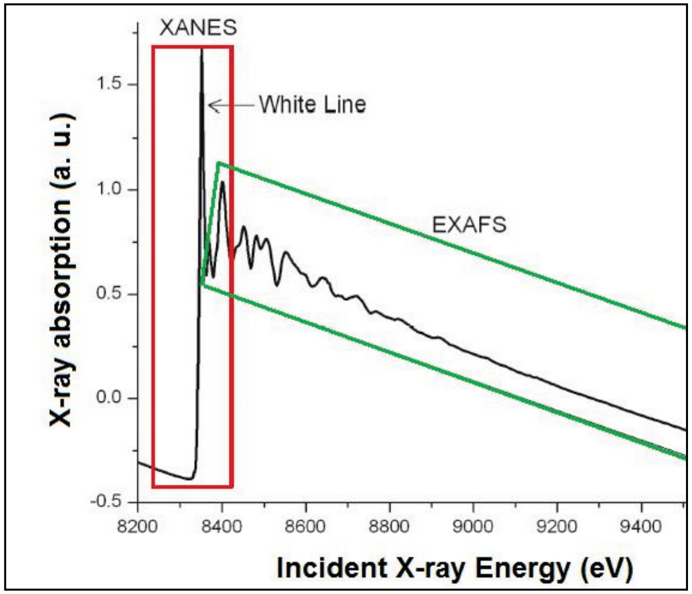
Example for $GaO_{1.2}$

Nature Materials Vol. 7, 391-398 (2008)

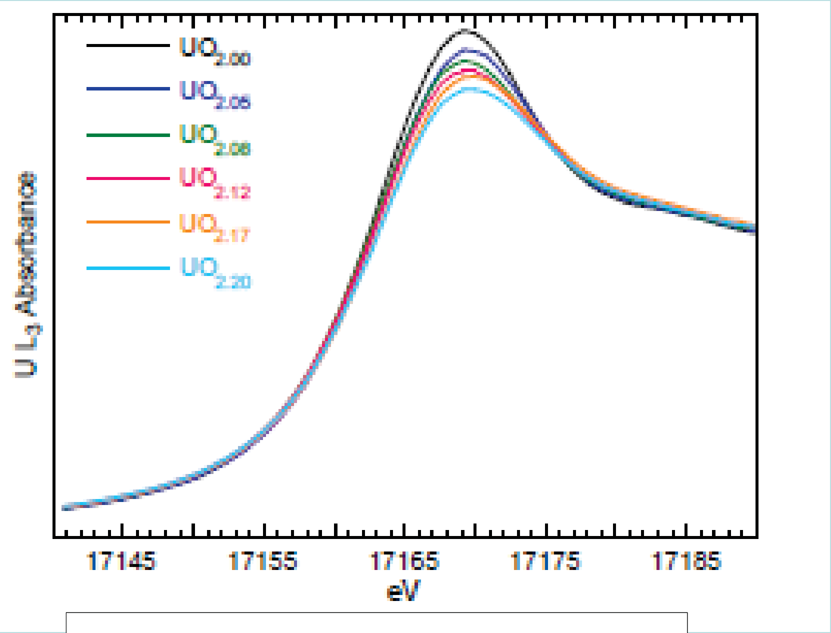
Explanation for X-Ray absorption techniques

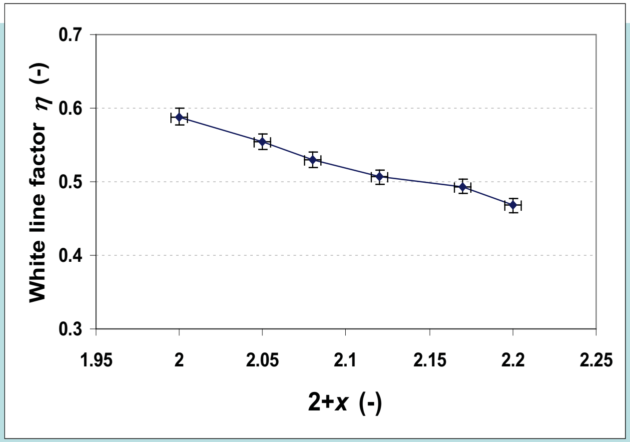
Effect of Redox on White line

White line



Example for UO_{2+x}





Conradson et al. *J. Solid State Chem.* 178 (2005) 521-535.

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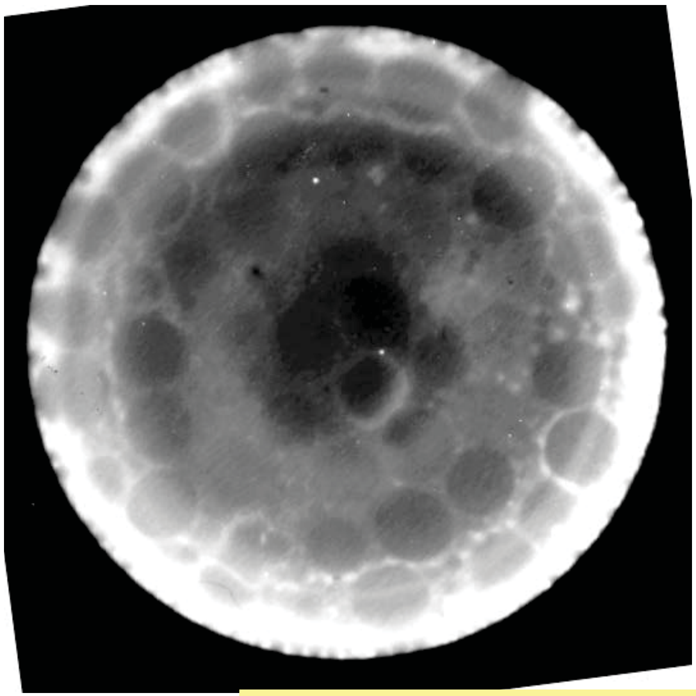
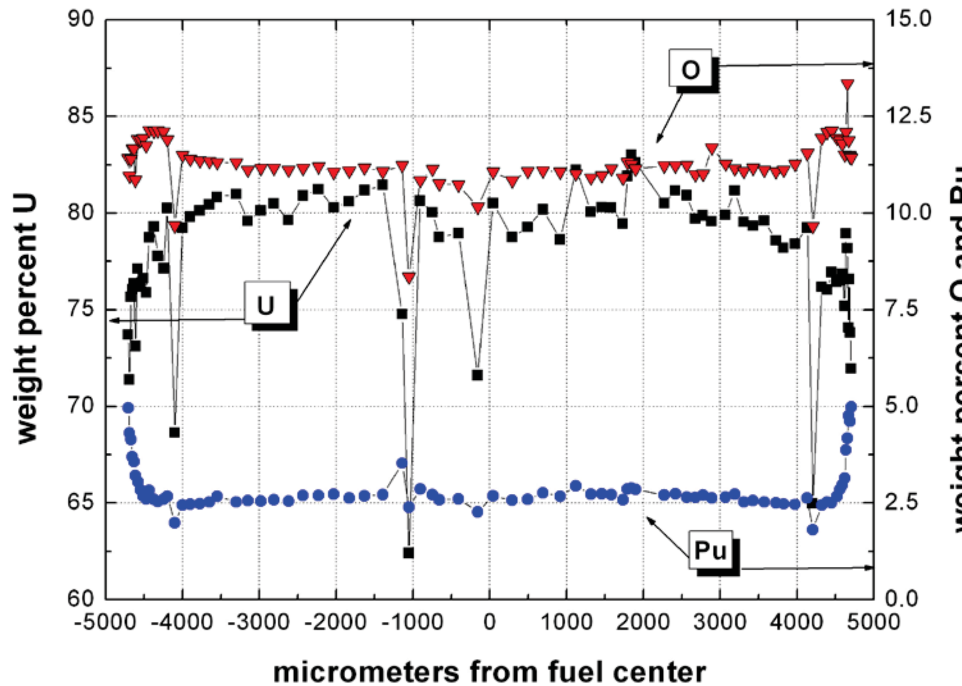
Example for µXRD and EXAFS - Sample

MOX sample: SpherePack 60 MWd kg-1 @ KKB-I

Beta and gamma autoradiography done on the sample





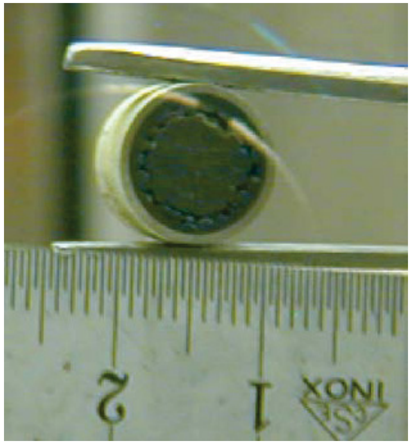
C. Degueldre*, M.A. Pouchon, G. Kuri, C. Borca, C. Cozzo, White line of actinide x-ray absorption spectra as a tool for their atomic environment description, 40emes Journées des actinides , CERN, 27-30 March 2010

We also have: diametrical concentration profiles of the fission products Nd, Zr, Cs, Xe, Ba, Mo and Ru.

C. Degueldre et al., J. Nucl. Mater. (2011), doi:10.1016/j.jnucmat.2010.11.096

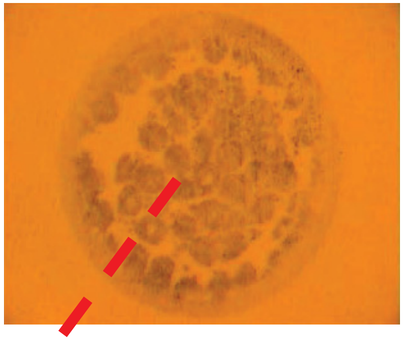
Example for µXRD and EXAFS - Setup

MOX XAFS analysis

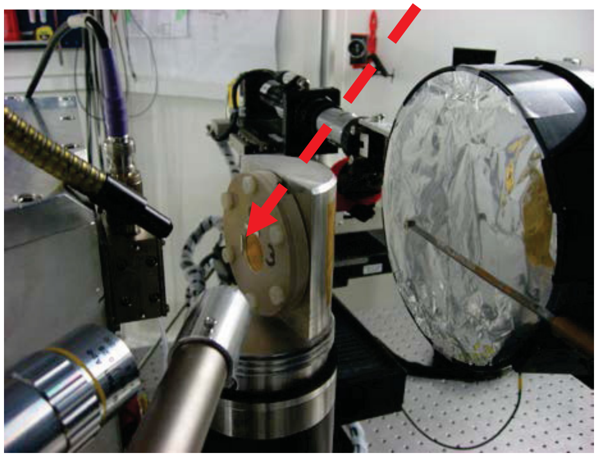


peeling and selection of **subsample** with activity **< 100 LA**

Kapton foil



µXAS setup



C. Degueldre et al., J. Nucl. Mater. (2011), doi:10.1016/j.jnucmat.2010.11.096

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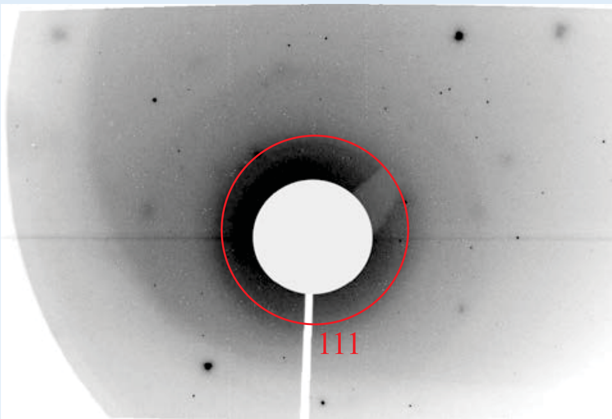
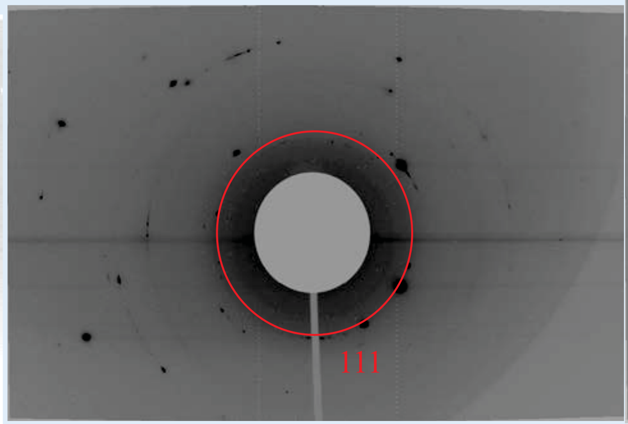
Example for µXRD and EXAFS - µXRD

MOX µXRD conditions: E_0 18600 eV, $\lambda = 0.666$ Å, $v \sim 300\ \mu m^3$

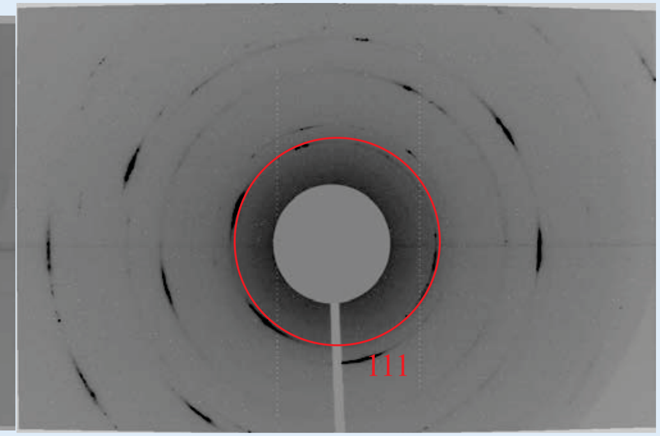
Pristine MOX



MOX center



MOX RIM



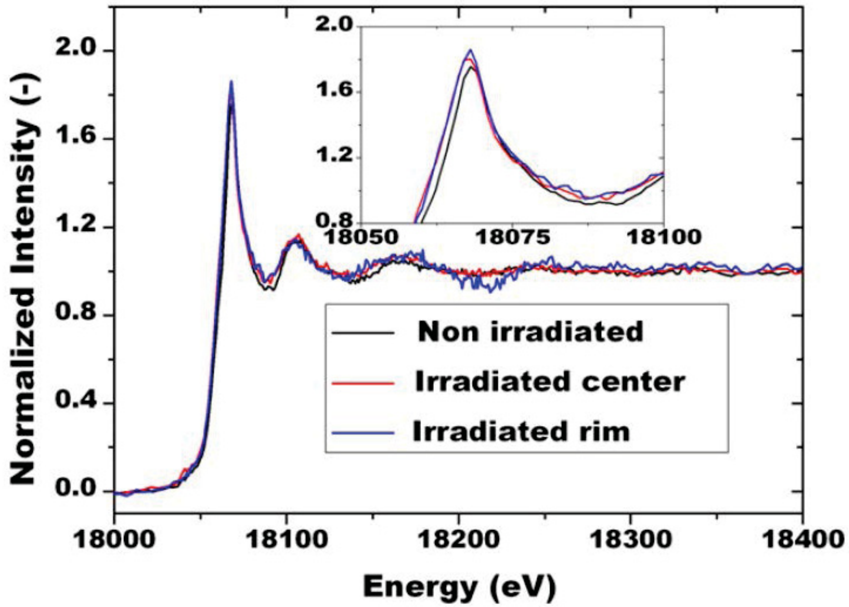
Local **crystalline changes during burnup**

- Center: lower burnup (1700dpa) **limits damage** & high T (~1400oC) **increases healing**
- RIM: larger burnup (2500dpa) **increases damage** & lower T (~600oC) **limits healing**

C. Degueldre et al., J. Nucl. Mater. (2011), doi:10.1016/j.jnucmat.2010.11.096

Example for µXRD and EXAFS – White line

MOX µXAFS conditions: Pu L3 Edge, *v* ~300 µm^3



Pu redox in MOX fuel is likely to remain unchanged during burnup

In MOX center, lower burnup (*E*o = 18062±2 eV).

In MOX RIM, larger burnup but PCI, (*E*o = 18062±2 eV).

C. Degueldre et al., J. Nucl. Mater. (2011), doi:10.1016/j.jnucmat.2010.11.096

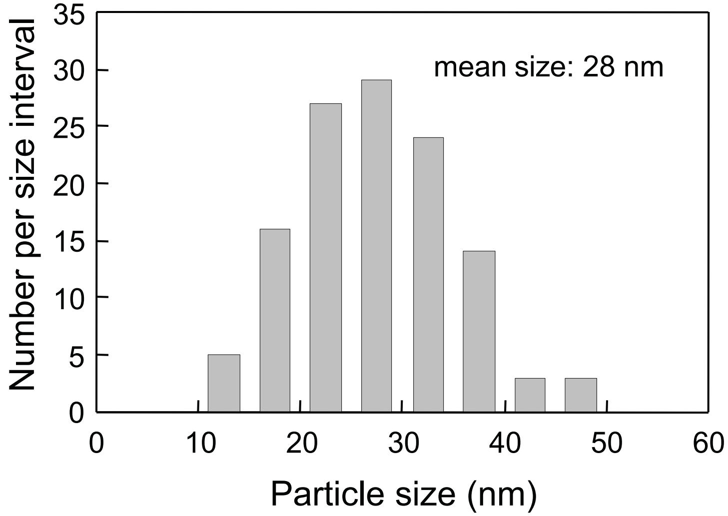
Example for EXAFS

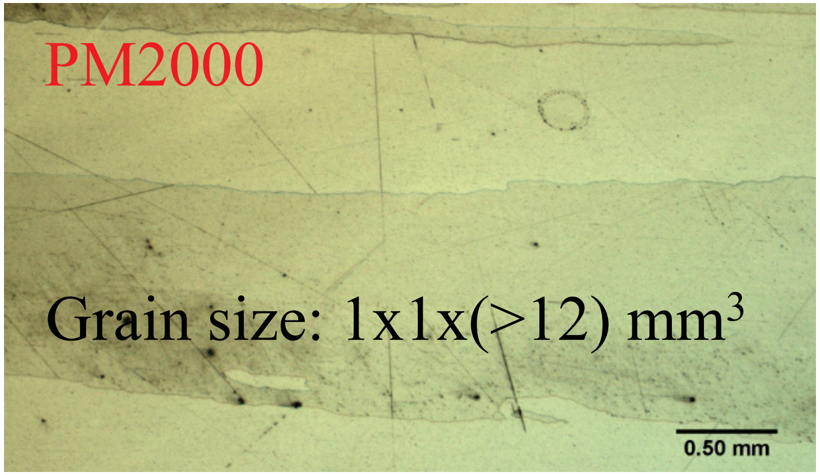
EXAFS of irradiated **ODS - Material**

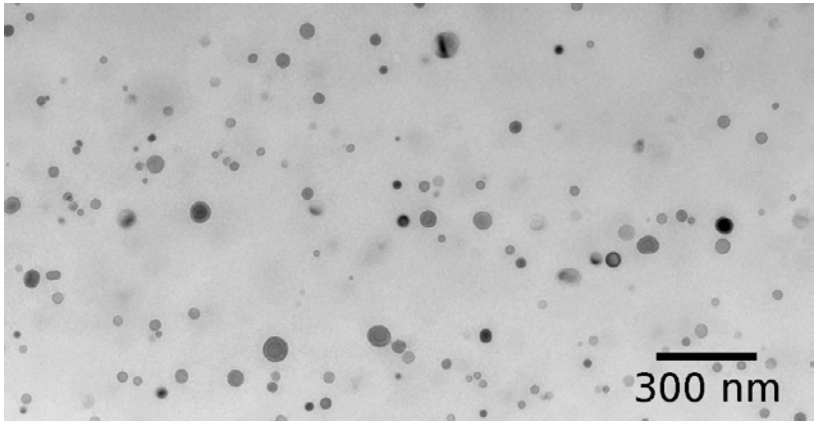
Chemical composition

elements	Fe	Cr	Al	Ti	Y_2O_3
wt %	73.5	20	5.5	0.5	0.5

Y_2O_3 particle size distribution







Pouchon, 10.4028/www.scientific.net/MSF.561-565.1761

Example for EXAFS

EXAFS of irradiated **ODS** - Cyclotron @ 570 K - Y_2O_3 Results

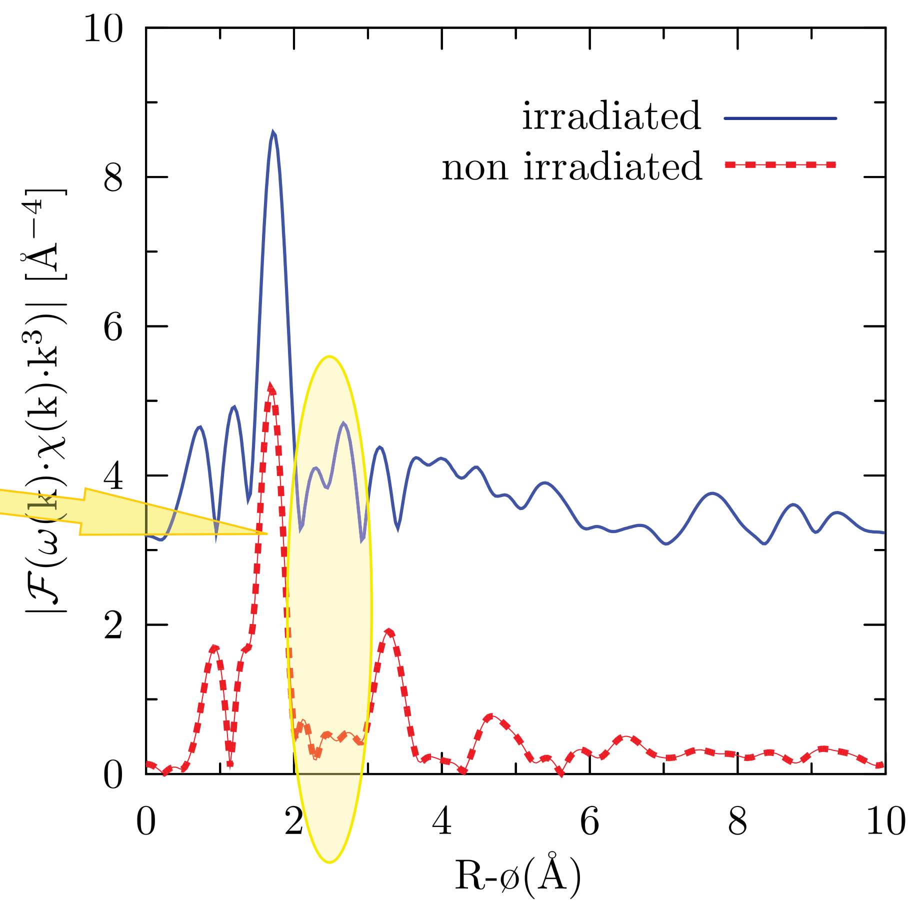
Comparison of the **EXAFS** signal for the **irradiated** and the **non-irradiated** sample

Difference!

Irradiation:

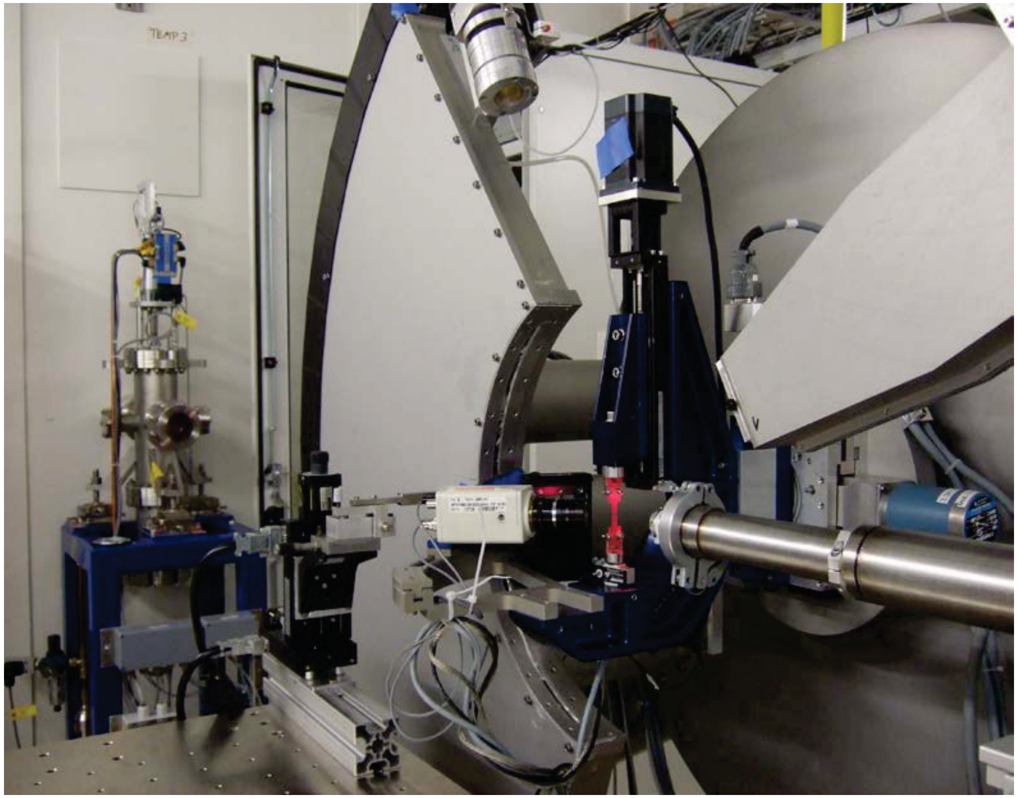
- 1 dpa
- 570 K irrad. temperature

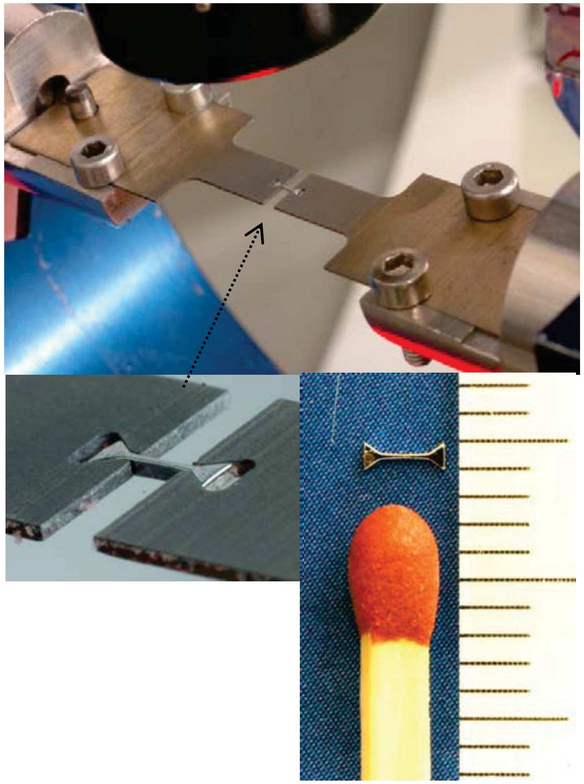
→ **Structure of Y_2O_3 changes**



Pouchon, 10.4028/www.scientific.net/MSF.561-565.1761

Example for in situ µ-XRD

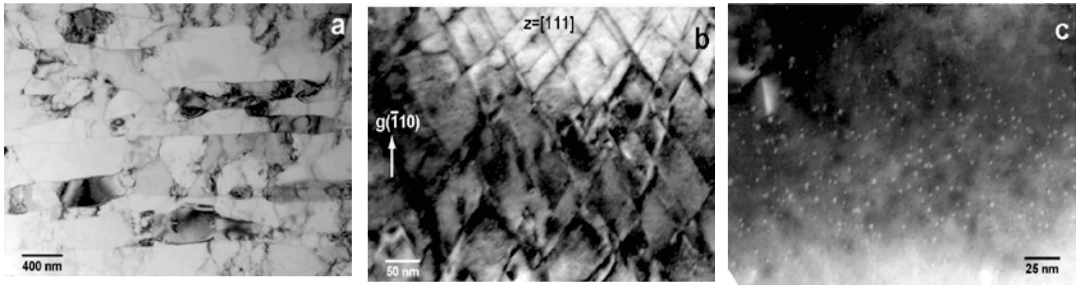


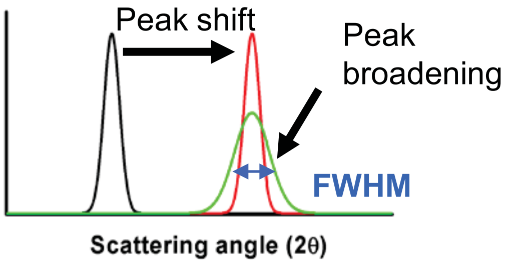


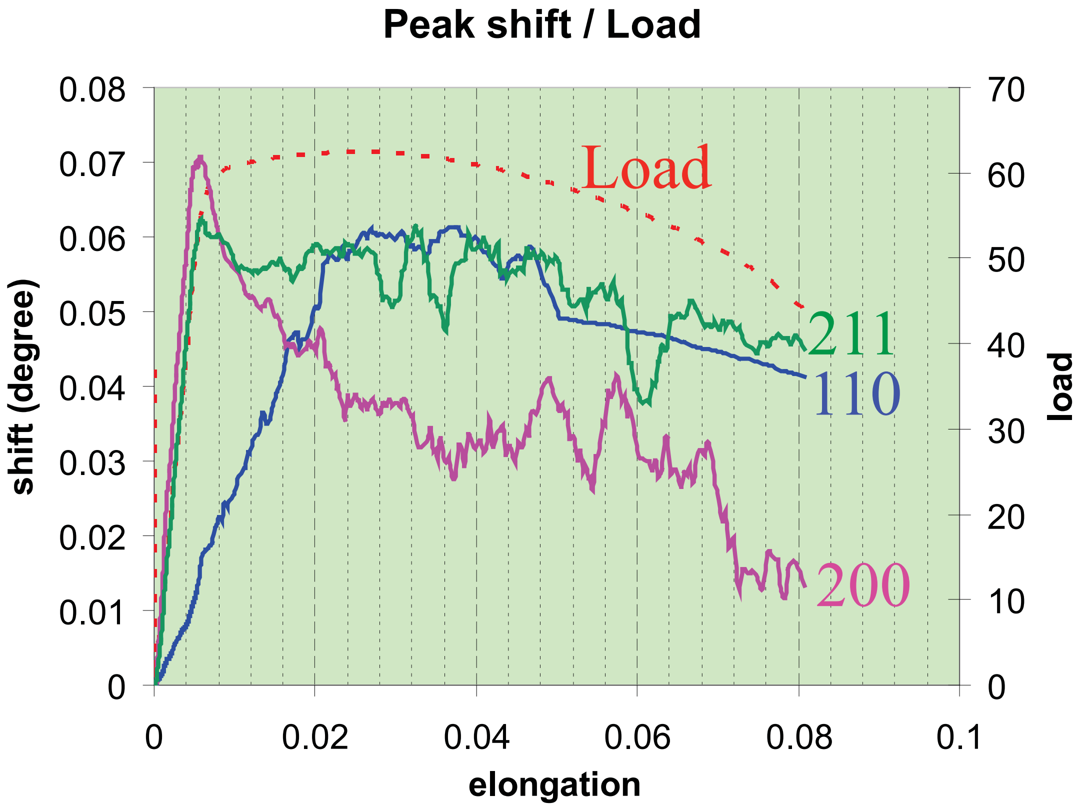
J. Zimmermann et al., In situ X-ray Diffraction Synchrotron Study of an Advanced ODS Ferritic Steel during Tensile Deformation

Example for in situ μ-XRD

ODS Material – K1 from Kyoto university







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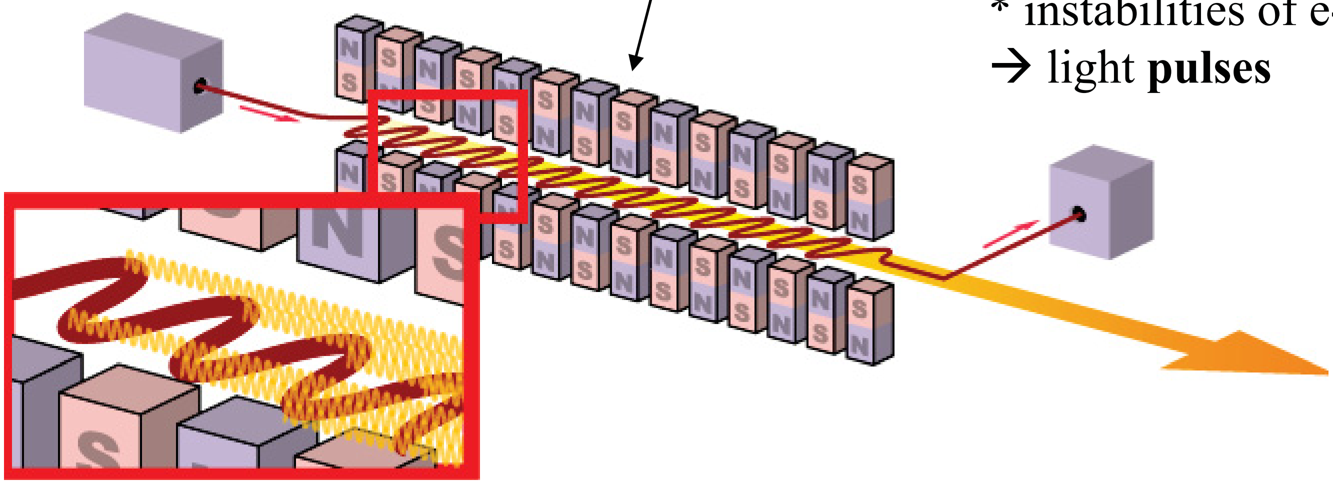
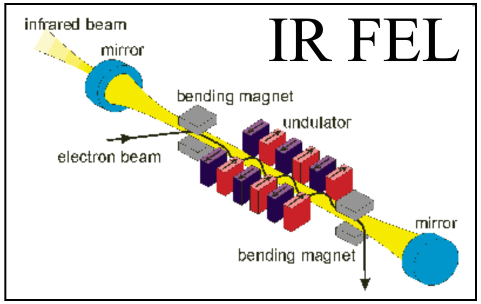
X-Ray FEL

undulator (wiggler) → light emission
* electron motion is in phase
with the field of the light already emitted
→ **coherency**
* instabilities of e- beam lead to bunching
→ light **pulses**



- **FLASH** in Hamburg
- **LCLS** at SLAC National Accelerator Laboratory
- **XFEL** in Hamburg
- **SCSS** at SPring-8
- **SwissFEL** at the PSI
  SACLA at the RIKEN Harima Inst.

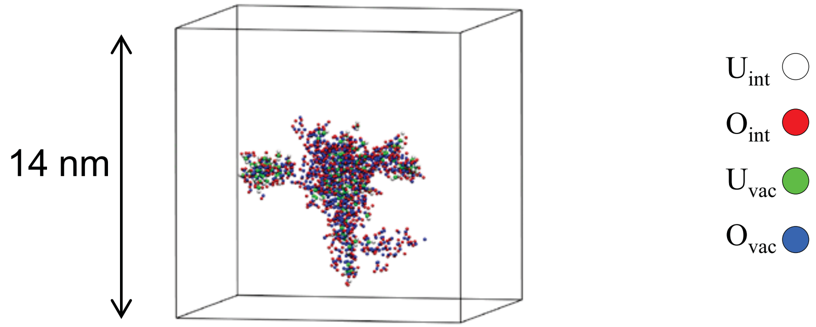
Laser:
- **coherent** electromagnetic radiation
- high **intensity**
- **pulsed** light (~30 fs with 10^{11} photons)



Possible application of a FEL

Taking advantage of very short (fs range) pulses:

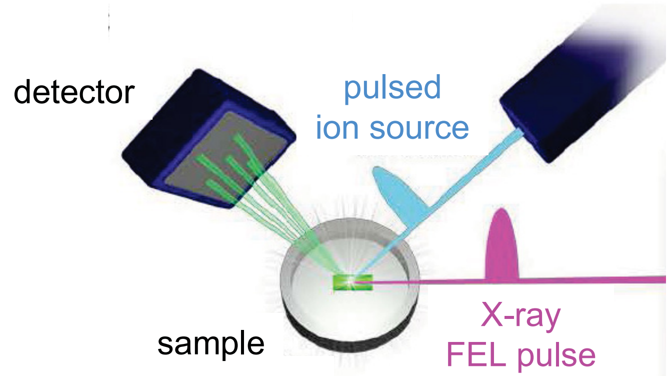
- Dynamics of irradiation-induced defects in nuclear materials



(from F. Devynck & M. Krack (NES / LRS))

t: ~ 0.1 - 30 ps, L: 0.1 - 10 nm

- Method: pump - probe diffuse scattering



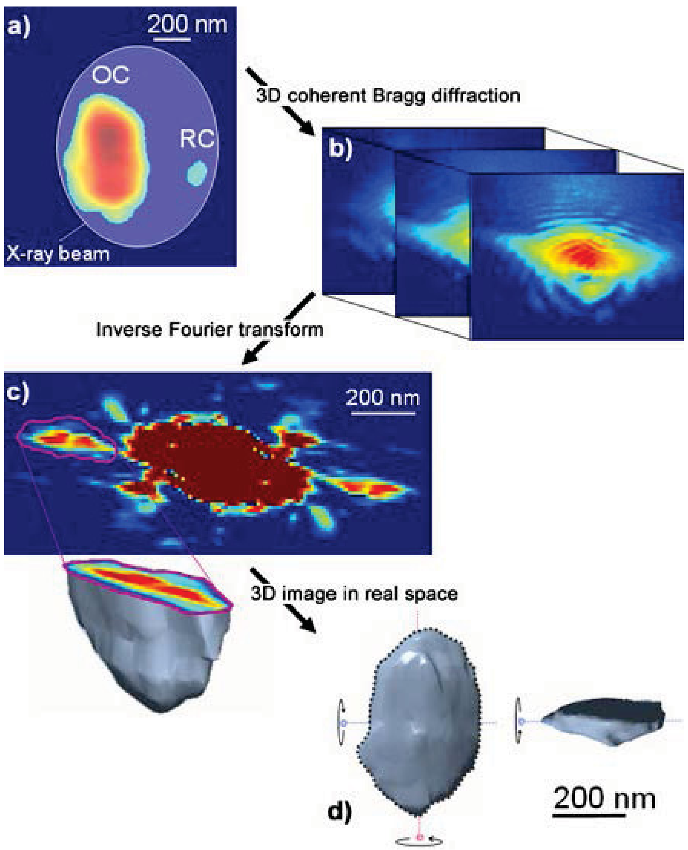
(from B. Larson, ORNL)

A. Froideval, A. Badillo, J. Bertsch, S. Churakov, R. Dähn, C. Degueldre, T. Lind, D. Paladino, B.D. Patterson, J. Nucl. Mater. (2011), article in press

Possible application of a FEL

Taking advantage of coherency:

→ Possibility to take advantage of holographic techniques



http://www.esrf.eu/news/spotlight/spotlight122

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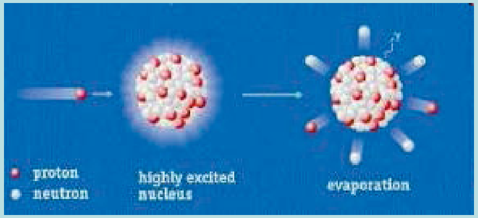
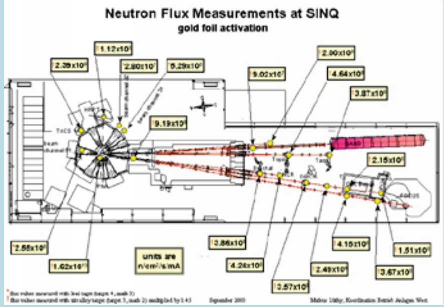
    - Selection of analysis techniques
    - Examples
      - PIE of MOX by µXRD, EXAFS
      - XRD of ODS after He irradiation
  - Free electron laser (FEL)
    - Special aspects/Possible application
- Neutrons
  - Sources / Features
  - One example

Neutron radiation – source - property

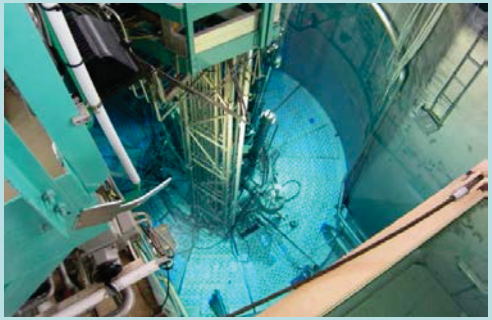
SOURCE

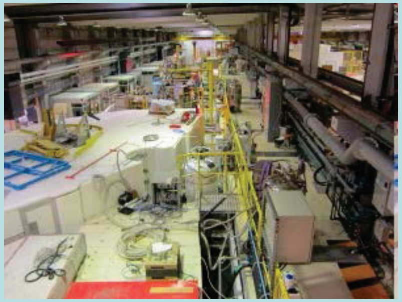
Spallation





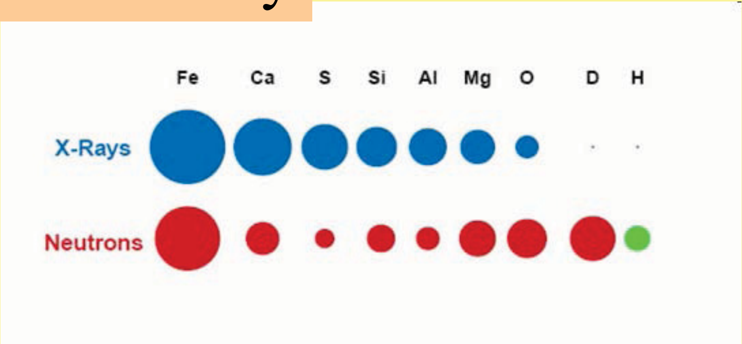
Nuclear reactor



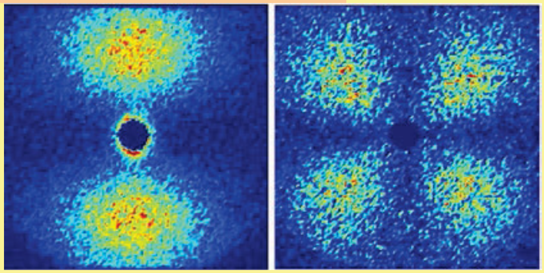


Features

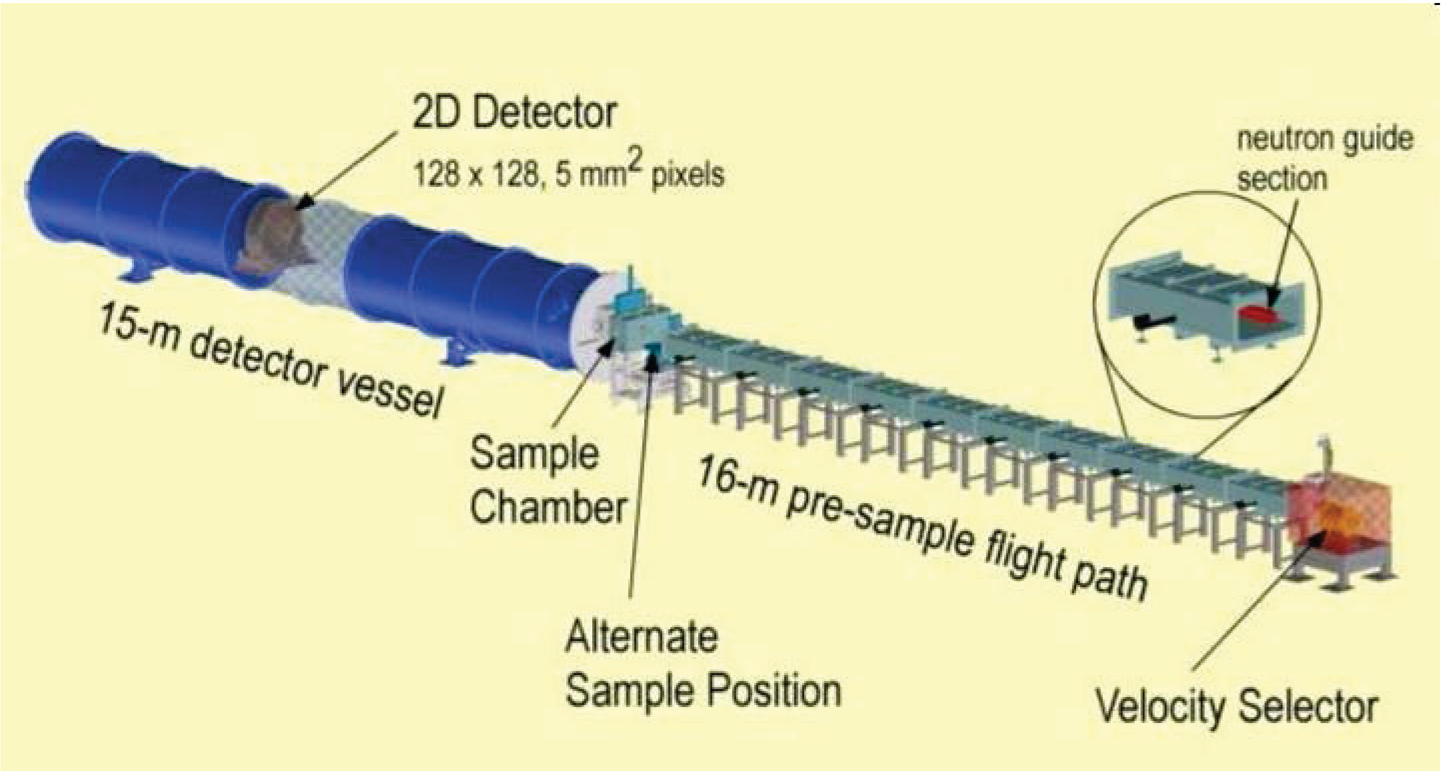
Sensitivity



Magnetic interaction



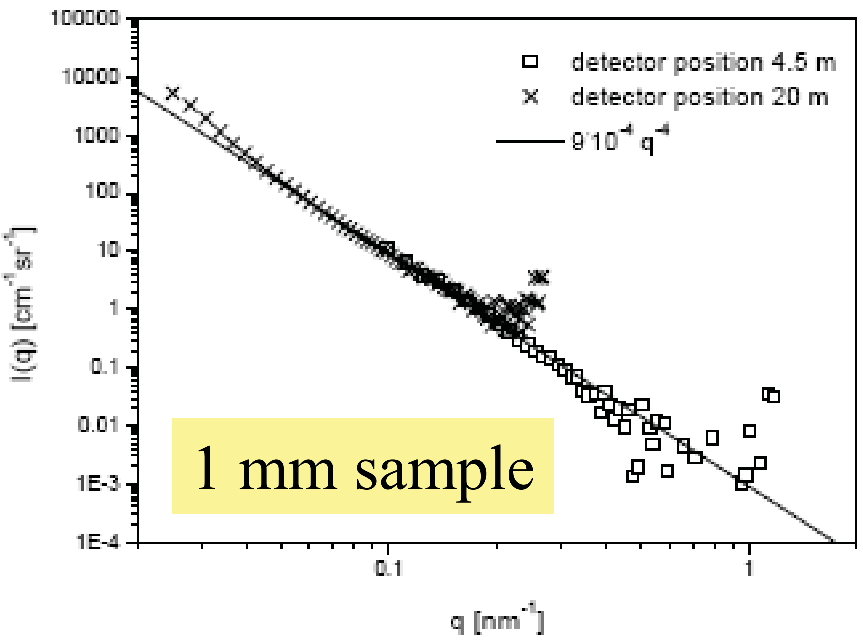
Example: small angle neuton scattering

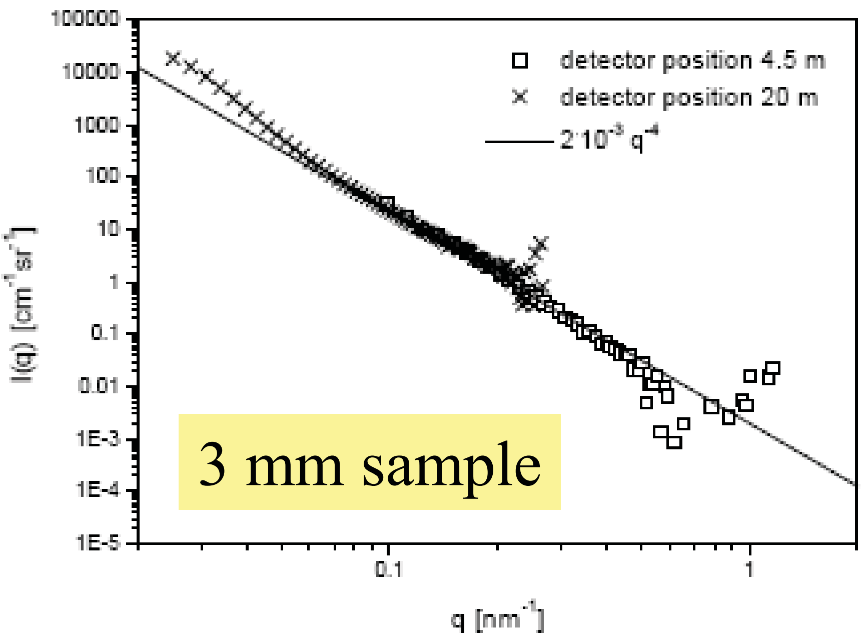


Detection of very small features

Example for Small angle scattering

Porosity investigation of a ceramic (YSZ) as an inert matrix fuel





→ Volumetric identification of porosity in the <300 nm range

http://sinq.web.psi.ch/sinq/er/ii_98s_55.pdf

Conclusions

- Beamline techniques offer many new possibilities, but require a good planning
- Radioactive samples will often require important downsizing / miniaturization and equipped beamlines
- Some techniques allow atomistic resolution, fast processes, …
  → useful for validation of modeling

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Laser-based Measurement Techniques for Nuclear Materials

J. Rory Kennedy, David Hurley, Robert Schley, Stephen Reese, Matthew Fig

Idaho National Laboratory

Idaho National Laboratory

OECD/NEA PIE Workshop

Paris, France

June 16, 2011

Topics

- **Thermal Diffusivity**
- **Laser-based Resonant Ultrasound**
  - Average elastic constants
  - Ultrasonic attenuation and microstructure
  - Dispersion relation
  - In-pile measurements

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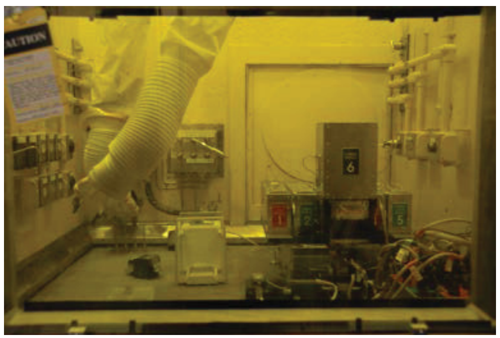
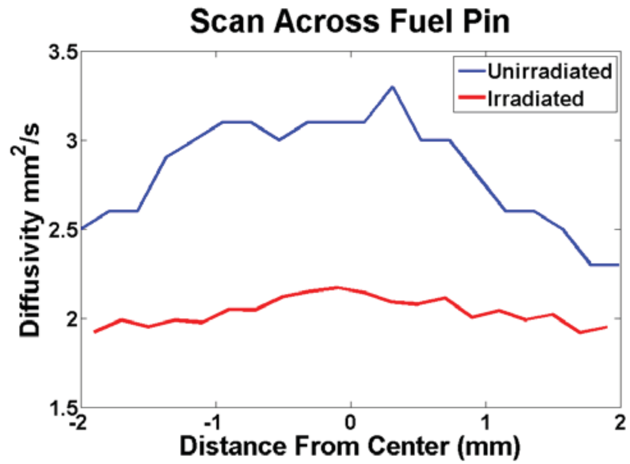
Characterization and Properties of Fuels:

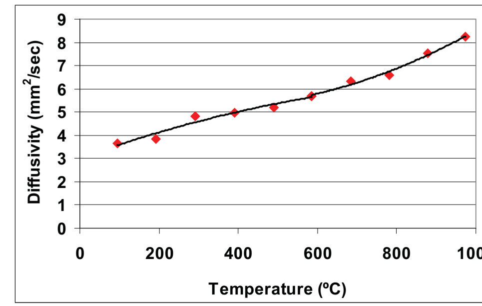
First radial profile of thermal diffusivity on FCRD/AFCI fuel samples (irradiated and unirradiated) using STDM

Scanning thermal diffusivity microscope (STDM) allows determination of thermal diffusivity at 50 µm spatial resolution (highest spatial resolution capability in world). Laser based and developed for hot cell application over past 4 years.

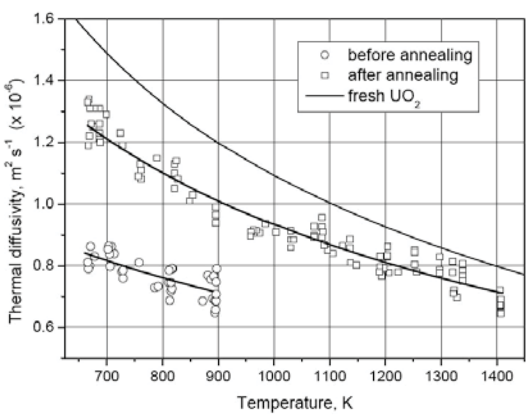


STDM in hot cell



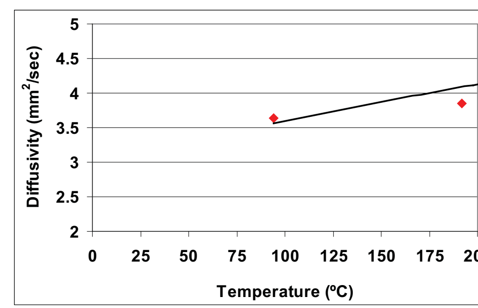


- Thermal diffusivity of bulk material from laser flash measurements.



D. Staicu, M. Joergensen, G. Pagliosa, D. Papaioannou, M. Sheindlin, C.T. Walker
ANS 2005 Water Reactor Fuel Performance Meeting, Kyoto, Oct. 2-6, 2005

- **Scan results at every 200 µm across diameter of 4mm metal fuel pin.**
- **Unirradiated: middle area of fuel yields thermal diffusivity of about 3.1 mm^2/sec dropping off at periphery to 2.3 – 2.5 mm^2/sec.**
- **Irradiated (~6% burnup): drop to ~2 mm^2/sec and flatter radial profile. Smaller decrease than in oxides.**



- Extrapolation of curve to room temperature yields value very close to 3.1 mm^2/sec.

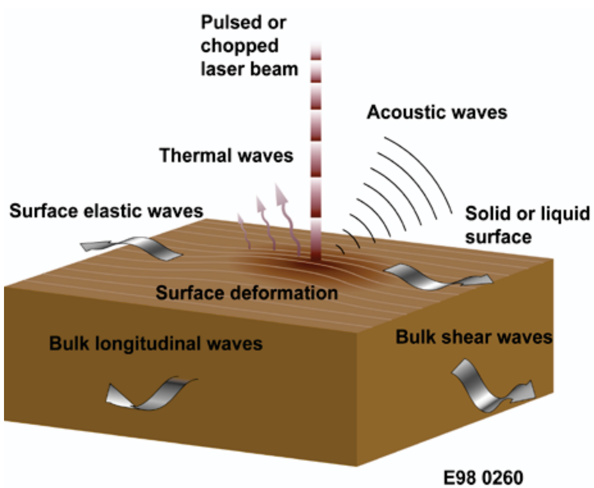
Laser-based Resonant Ultrasound: An overview

Laser Ultrasonics

Thermoelastic generation

Laser interferometric detection

Ideal for remote sensing applications



Resonant Ultrasound Spectroscopy

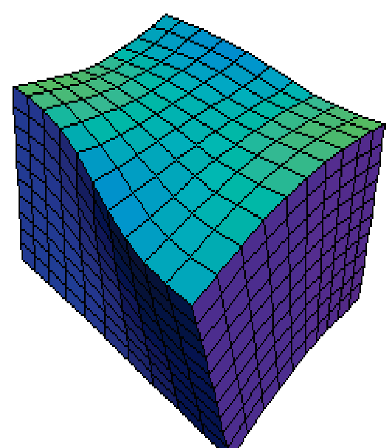
Effective tool to measure mechanical properties of materials, which are function of microstructure

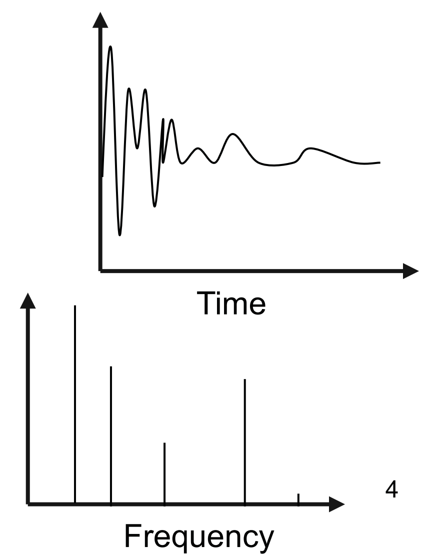
Relate signal to elastic stiffness tensor

Relate attenuation to anelastic behavior

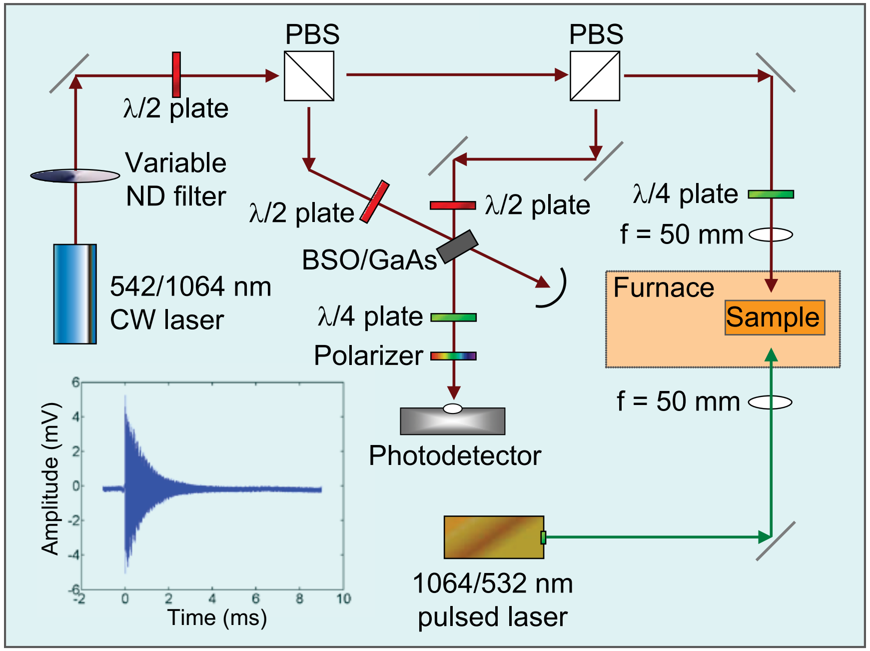
Typically use contact transducers

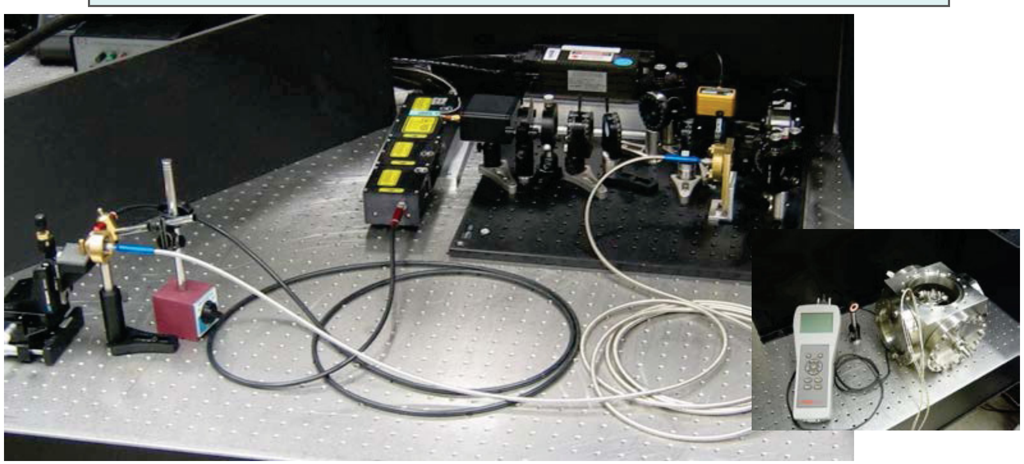
Typically use narrowband generation





Combining Laser Ultrasonics and Resonance Ultrasound Spectroscopy





- **Broadband resonant ultrasonic modes thermoelastically excited – generation and detection of all modes. Fast acquisition times → can see dynamic changes in microstructure mediated mechanical properties.**
- **Non-contact photorefractive interferometer detects out-of plane surface motion.**
- **Non-contact – more pure attenuation measurement.**
- **Raster scanning ability – allows mode identification.**
- **Remote operation – ideal for in-situ monitoring.**
- **Monitor microstructural evolution when combined with modern metallurgical imaging by tracking modes.**

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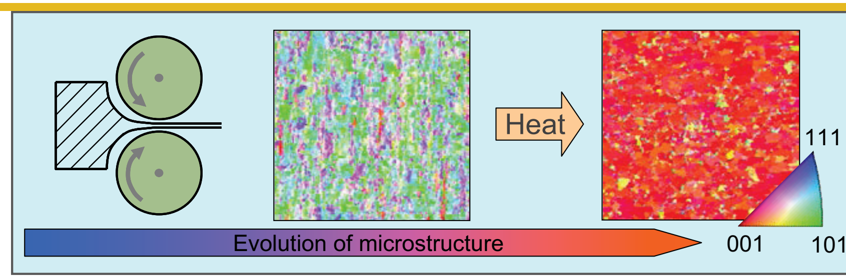
Characterization Technique Development

Demonstrated in-situ laser-based resonant ultrasound (LRUS) measurements of microstructure mediated mechanical property evolution

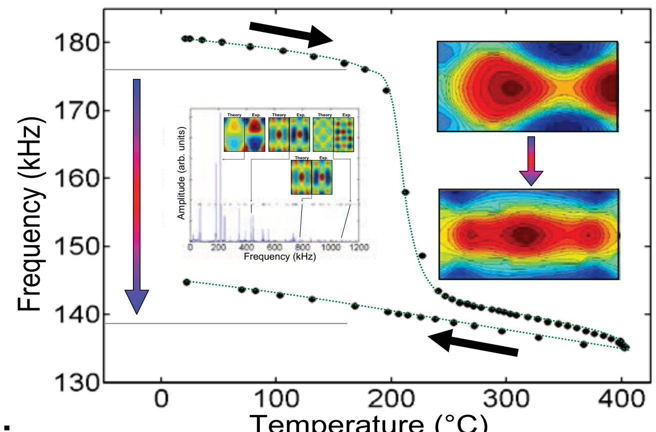
- High purity Cu can be cold rolled to produce a microstructure that changes dramatically upon heat treatment as observed using Electron Backscatter Diffraction (EBSD).



Electron Backscatter Diffraction (EBD)

- Non-contact LRUS allows generation and detection of all resonance modes in one measurement. Since acquisition times are fast, dynamic changes in microstructure mediated mechanical properties can be detected.

- Raster scanning before and after annealing yields image of each resonant mode and allows tracking of individual modes during annealing. Theory connects mode shape to polycrystal average elastic stiffness tensor derived from EBSD data.



Non-contact LRUS

- Current work – directly correlate this experiment with modeling and simulation that can predict the relationship between the polycrystalline elastic constants and the evolving microstructure. This is accomplished by coupling a polycrystal plasticity model with a phase field simulation to capture the defect-driven grain growth.

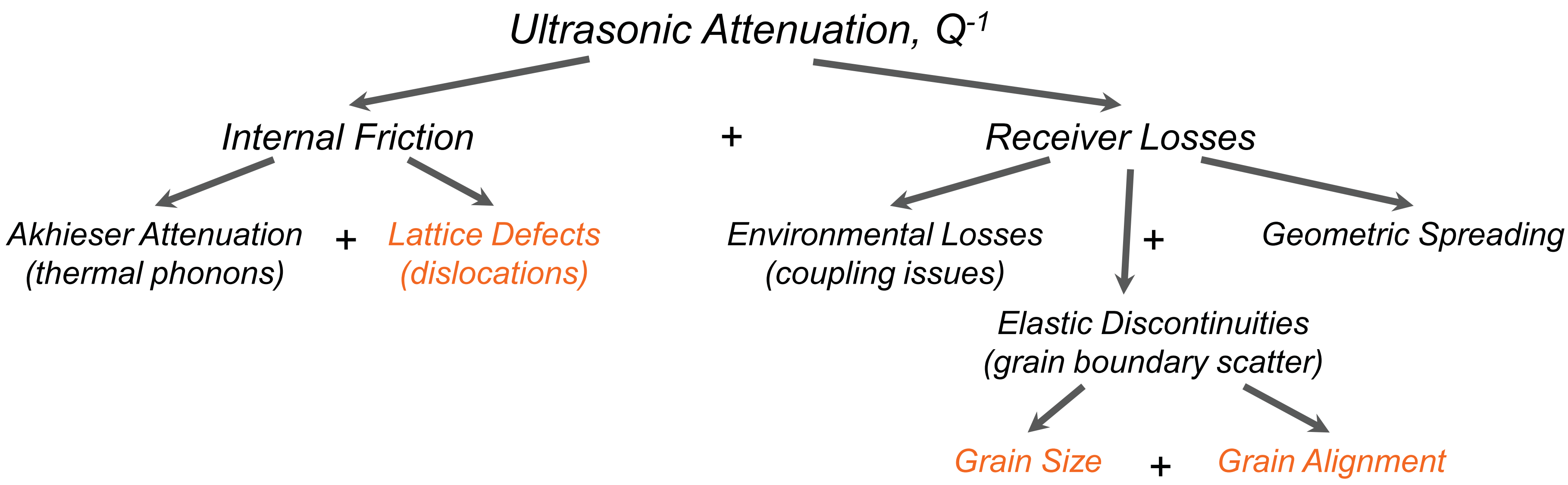
$$\overline{C}_{11}, \overline{C}_{12}, \overline{C}_{44}$$

Averaged elastic constants

- Averaged elastic constants are both experimentally and computationally determined as a function of time. RUS can provide a crucial validation metric for this modeling approach

Hurley, Reese, Park, Utegulov, Kennedy, Telschow J. Appl. Phys. 107, *2010*, (063510-) 1-5

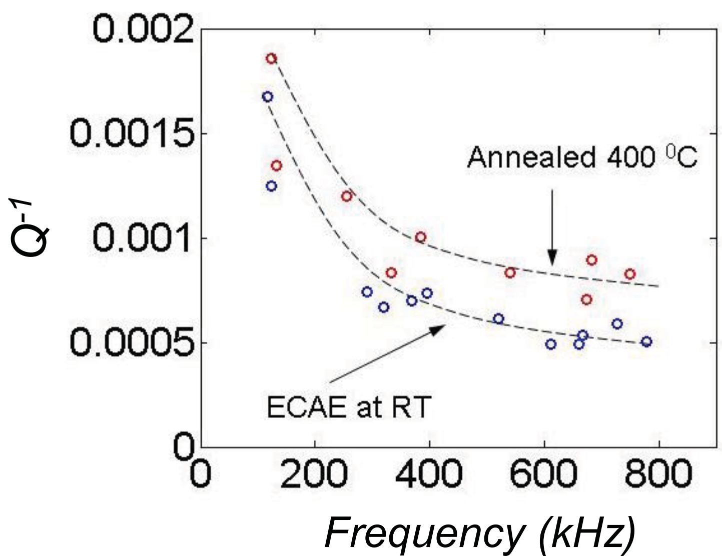
Laser-Based Material Characterization Techniques Development – Ultrasonic Attenuation

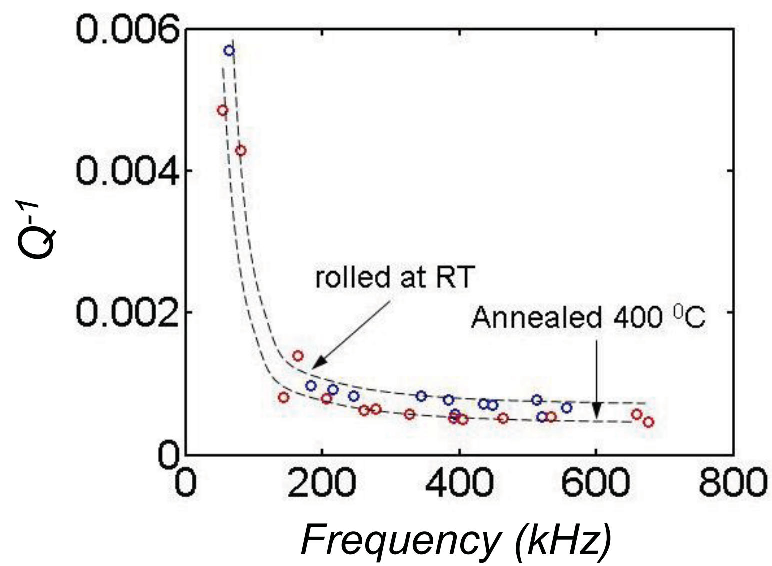


- **Akhieser attenuation is a function of temperature and can be known.**
- ***Environmental losses are minimized using a non-contacting laser-based measurement apparatus.***
- **Geometric spreading is irrelevant for measurements made in the frequency domain (i.e., only an issue with time-based measurements).**
- **Dislocations, grain size, and grain alignment are material properties governed by the state of the microstructure. Parsing their contributions to the attenuation measurement is key to accurately revealing the state of, or changes in, the material *microstructure.***

Laser-Based Material Characterization Techniques Development – Ultrasonic Attenuation

- **Equal channel angular extrusion (ECAE) sample lacks texture. During annealing, dislocations are annihilated and grains grow.**
- ***Rolled sample has strong texture. During annealing, dislocations are annihilated, grains grow and align.***
  - ***Decreasing dislocation density decreases attenutation (Q^{-1}).***
  - **Grain growth increases Q^{-1}.**
  - **Grain alignment decreases Q^{-1}.**





- ***In the ECAE sample, Q^{-1} increases overall during annealing (left figure), showing that grain growth is a larger factor than dislocation density.***
- ***In the rolled sample, Q^{-1} decreases overall during annealing (right figure), showing that grain alignment (+ dislocation density) is a larger factor than grain growth.***
- ***These two results combined show that grain boundary scatter (i.e., grain growth and alignment) is the dominant attenuation mechanism (vs. dislocation density).***

Characterization of corrosion (surface)

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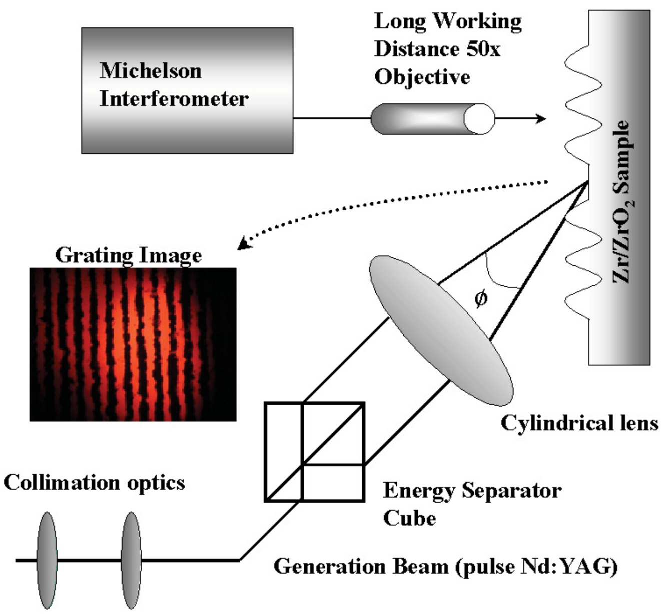
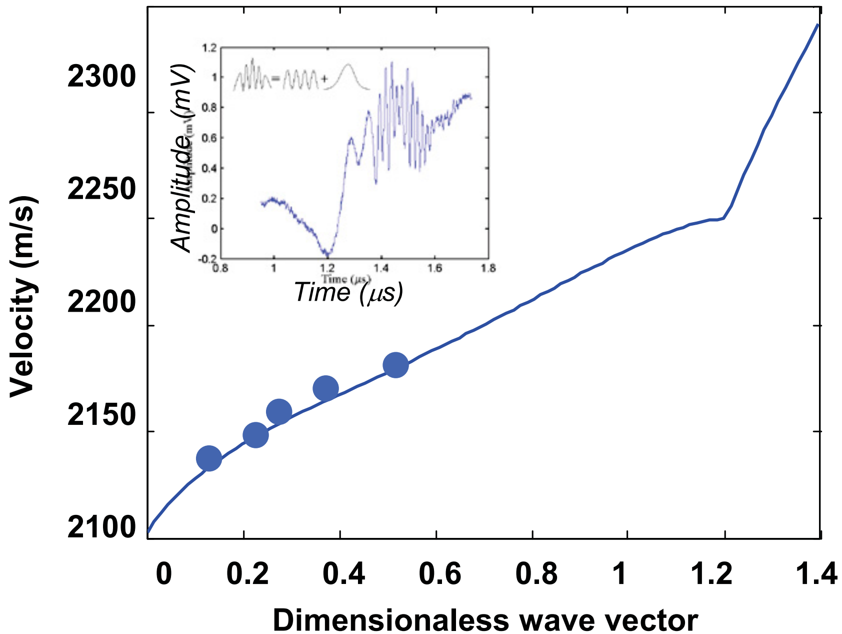
Experimental Setup (CW/pulsed)



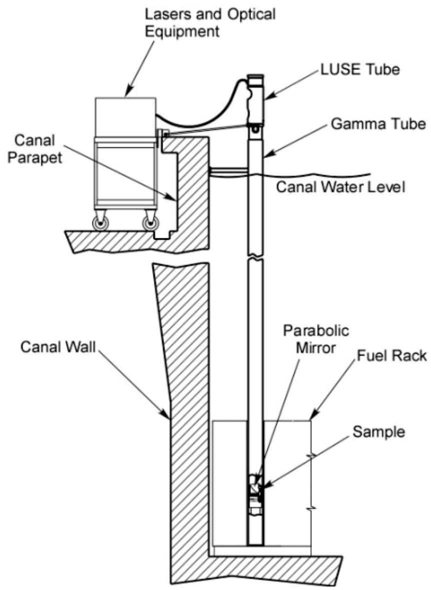
Dispersion relationship

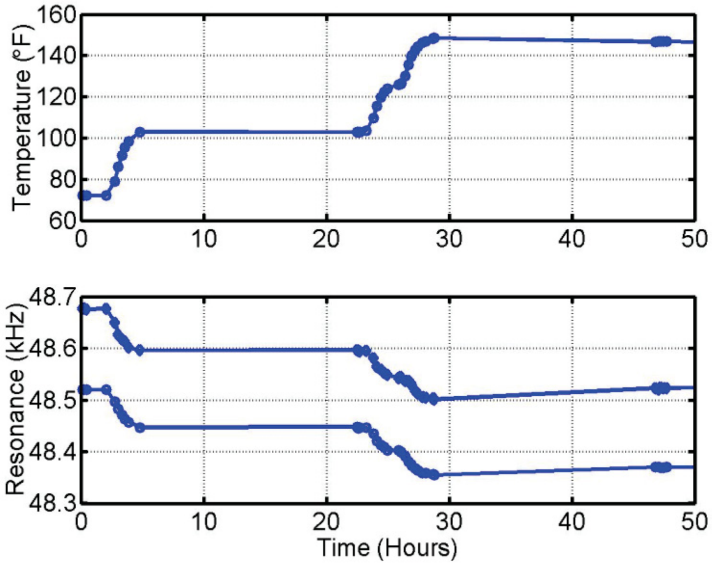


- Single frequency generation (CW) involves setting up an absorption grating at the samples surface by interfering two laser beams.
- This approach involves building the dispersion relation one point at a time. Dispersion relation can be used to back out the layer thickness
- Applications include monitoring the growth of corrosion films and hydride layers

In-pile measurements

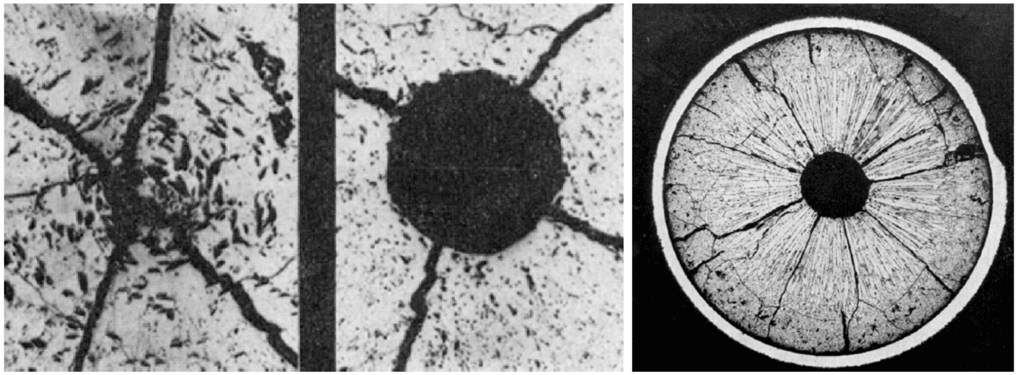
Laser RUS in high radiation environment: Gamma tube facility at ATR





In situ laser ultrasonic measurements of Inconel in the high gamma radiation field at ATR showing the sample temperature and resonant frequencies of a split vibration mode as irradiation was increased by (3 times) placing fuel rods closer to the sample.

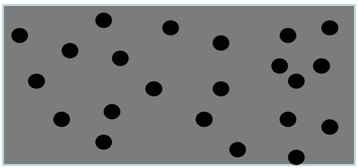
Central void formation in UO_2



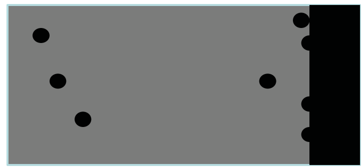
- The sintering process used to manufacture UO_2 fuel pellets results in a material with a density greater than 90% theoretical, with the porosity distributed uniformly throughout the pellet
- During the first day of reactor operation, the initial porosity migrates towards the center of the fuel pellet, eventually forming a center void
- In addition, a unique columnar grain structure with low porosity forms around the center void, with the grains oriented radially outward from the center
- A steep temperature gradient was applied to a UO_2 sintered fuel pellet out of pile and center void formation and columnar grain structure were observed (MacEwan and Lawson)

Central void formation in UO_2

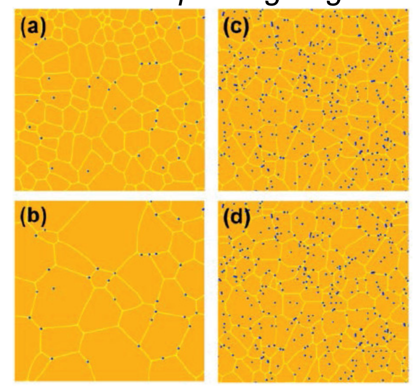
Initial Microstructure



Final Microstructure

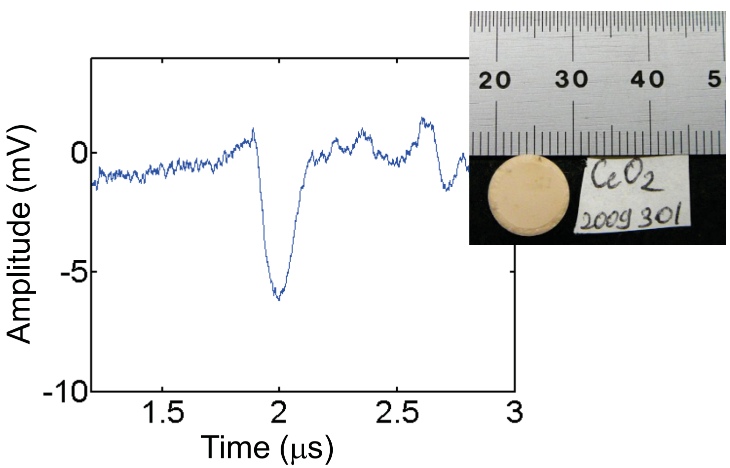


Void pinning of g.b.



- Link void pinning and void migration models
- As a void migrates up the temperature gradient, it drags the grain boundary forming a columnar structure

Measure spatial variation in porosity using laser ultrasound



For long wavelengths we can relate phase velocity to porosity using semi-empirical analytical formulas:

$E=E_0\exp(-bp)$

Duckworth, J. Amer. Ceram. Soc. 34 (1951) 1

Viscoelastic behavior for shorter wavelengths

X-Ray Microtomography

OECD/NEA International PIE Workshop

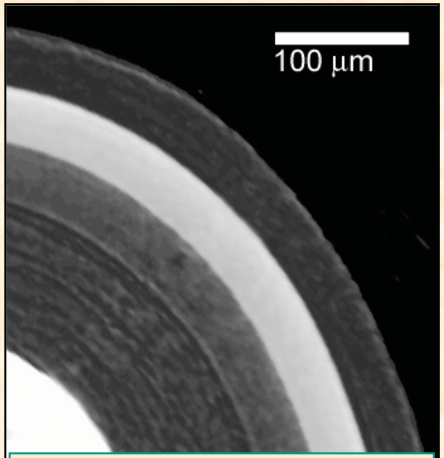
Paris, France

June 16, 2011

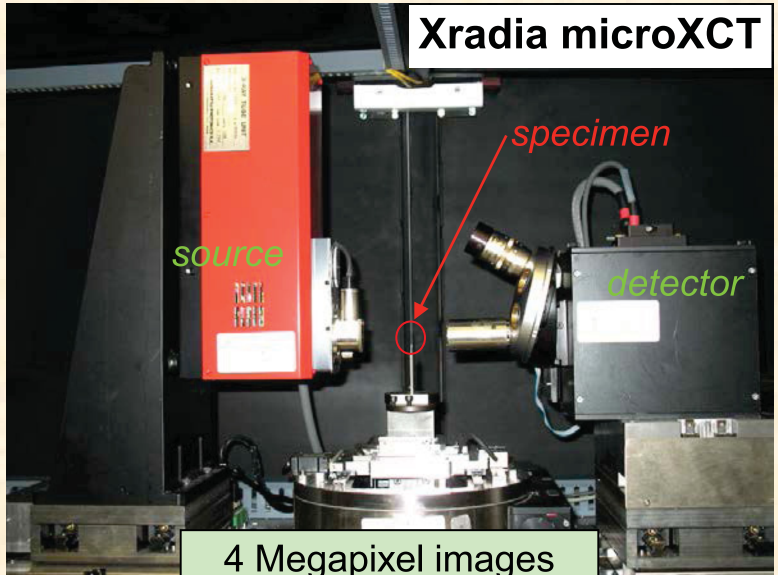
Lance Snead

Oak Ridge National Laboratory

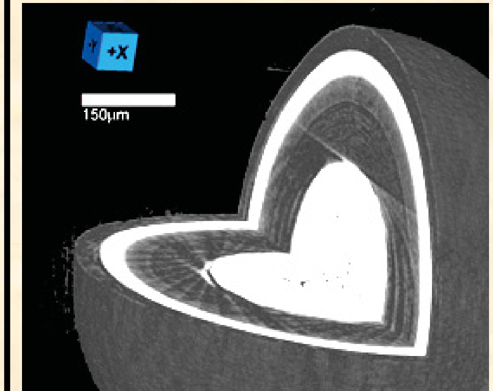
X-ray Microtomography



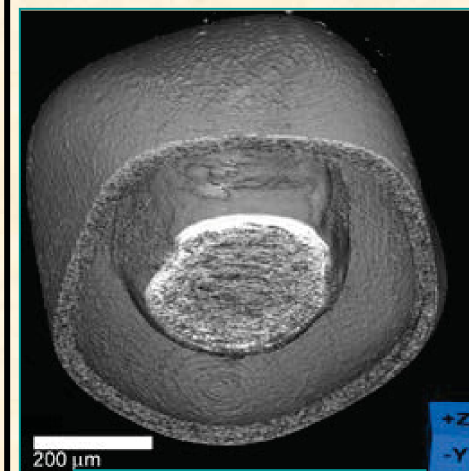
high resolution imaging



4 Megapixel images
1.2 mm field of view
1 - 2 µm resolution

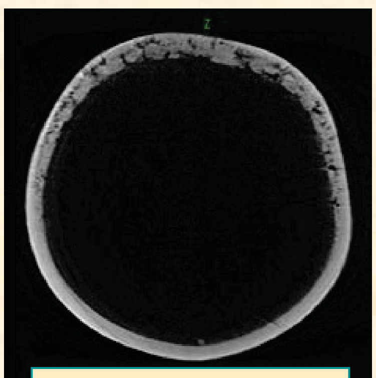


3-D visualization

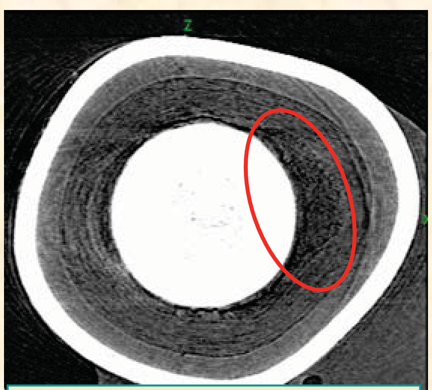


aspherical kernel

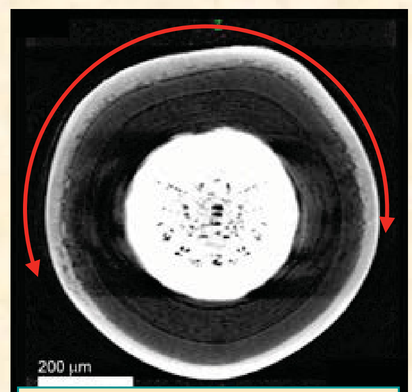
Nondestructive cross-sectioning



through thickness
SiC defect



soot inclusion in buffer



soot inclusion in SiC

X-ray Microtomography Application to Hot Samples

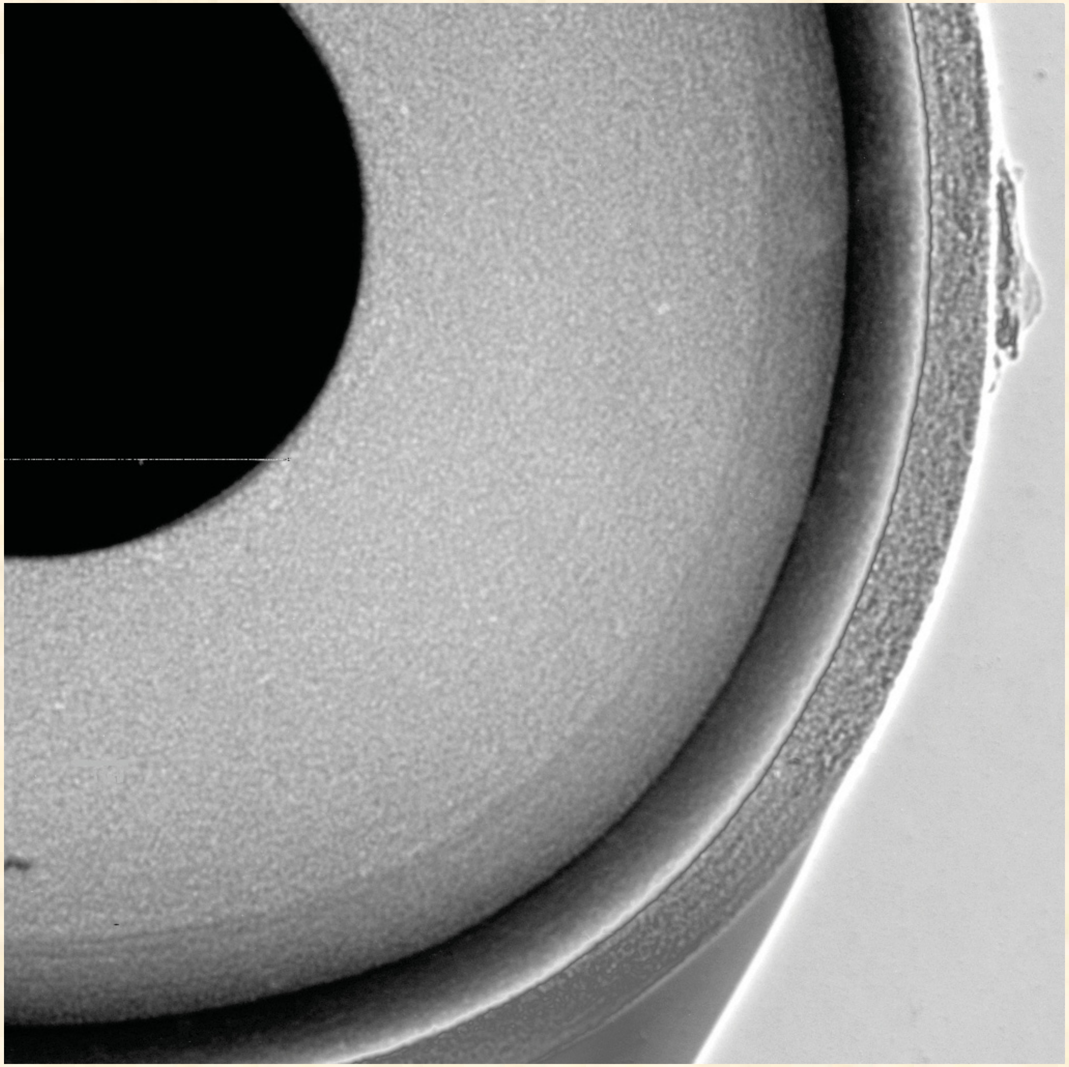
• Unit located in LAMDA facility (Low Activation Materials Development Analysis)

• Applied to irradiated graphite, surrogate and fueled TRISO (up to 100 mR/hr at one foot.)

• Current Upgrades Underway
  - Significant increase in resolution (sub-micron) and contrast and rad hard hardening optics.
  - Shielded transfer/loading for TRISO and other highly activated samples

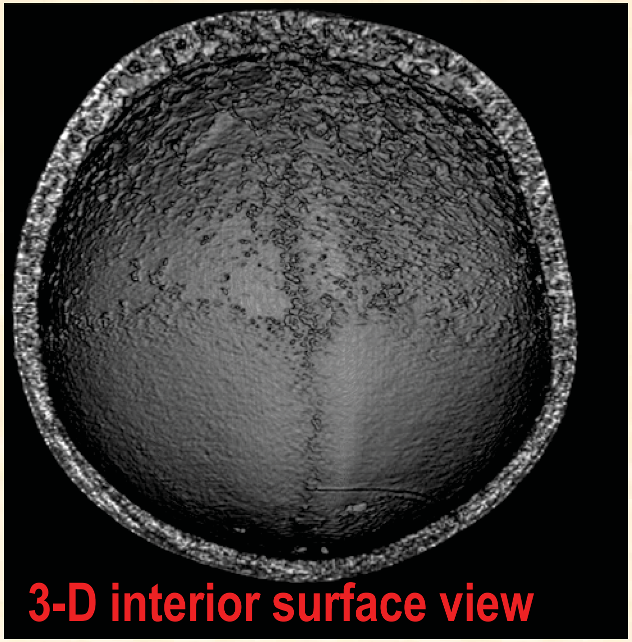
Actual Imaging Resolution of Current System
Surrogate TRISO Particle

1.4 µm
Resolution



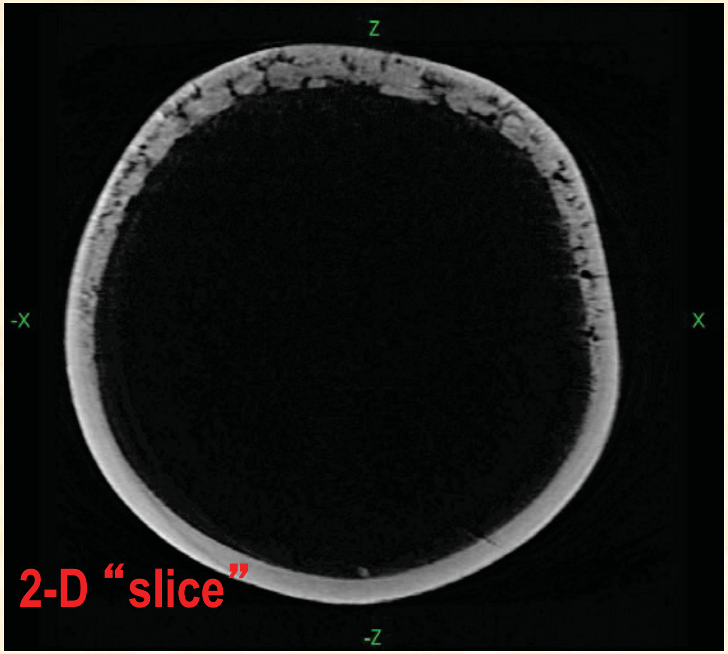
100 µm

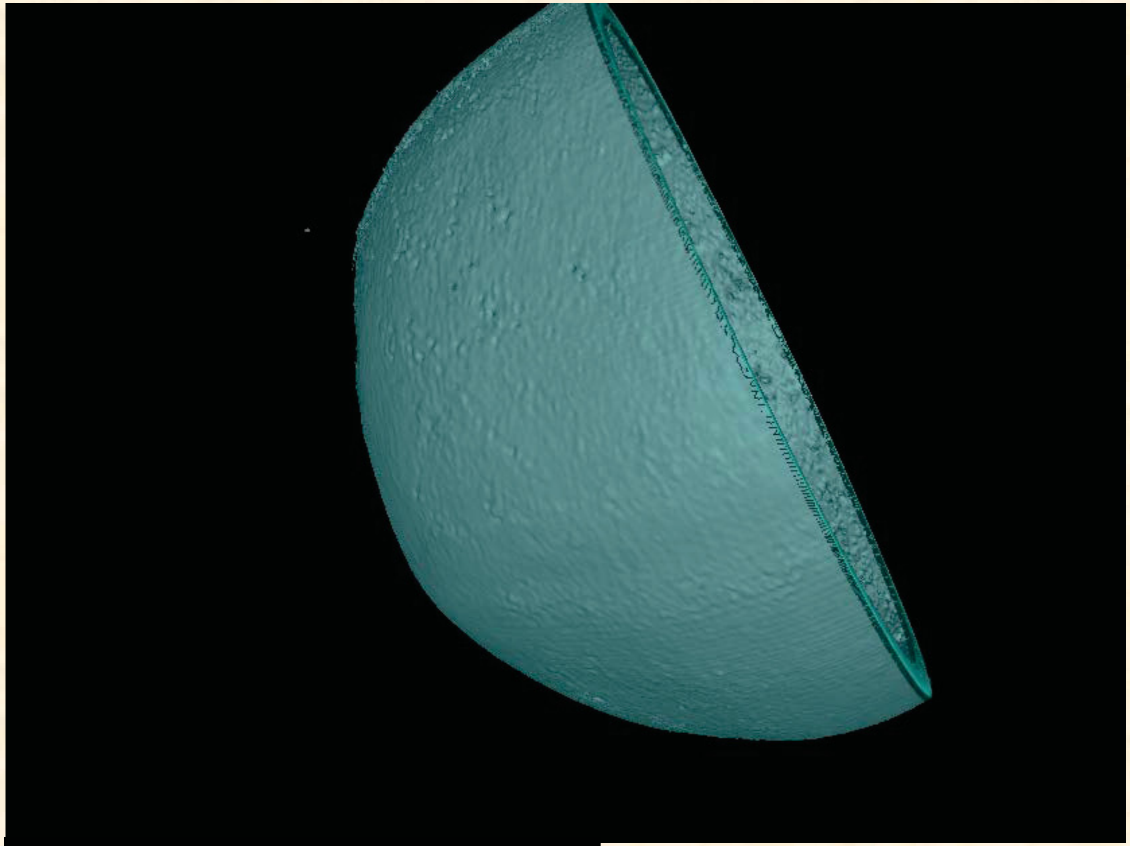
Capability of different visualizations of TRISO particle features



tomographs can provide 2-D slices, 3-D maps, and topographic views of x-ray density

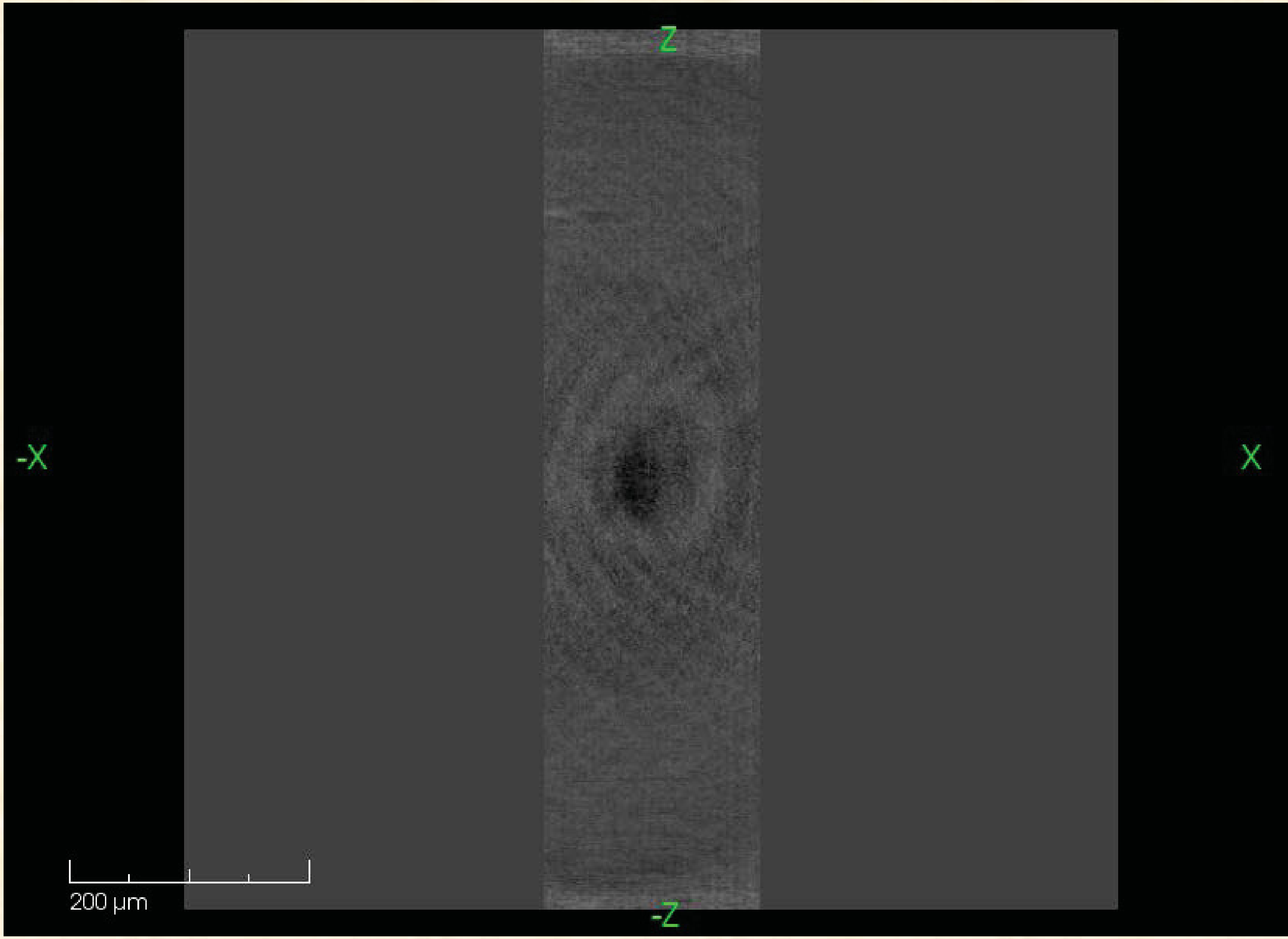
these tomographs show a hemispherical defect in the SiC layer





Animation of 3-D surfaces
(defective hemisphere)

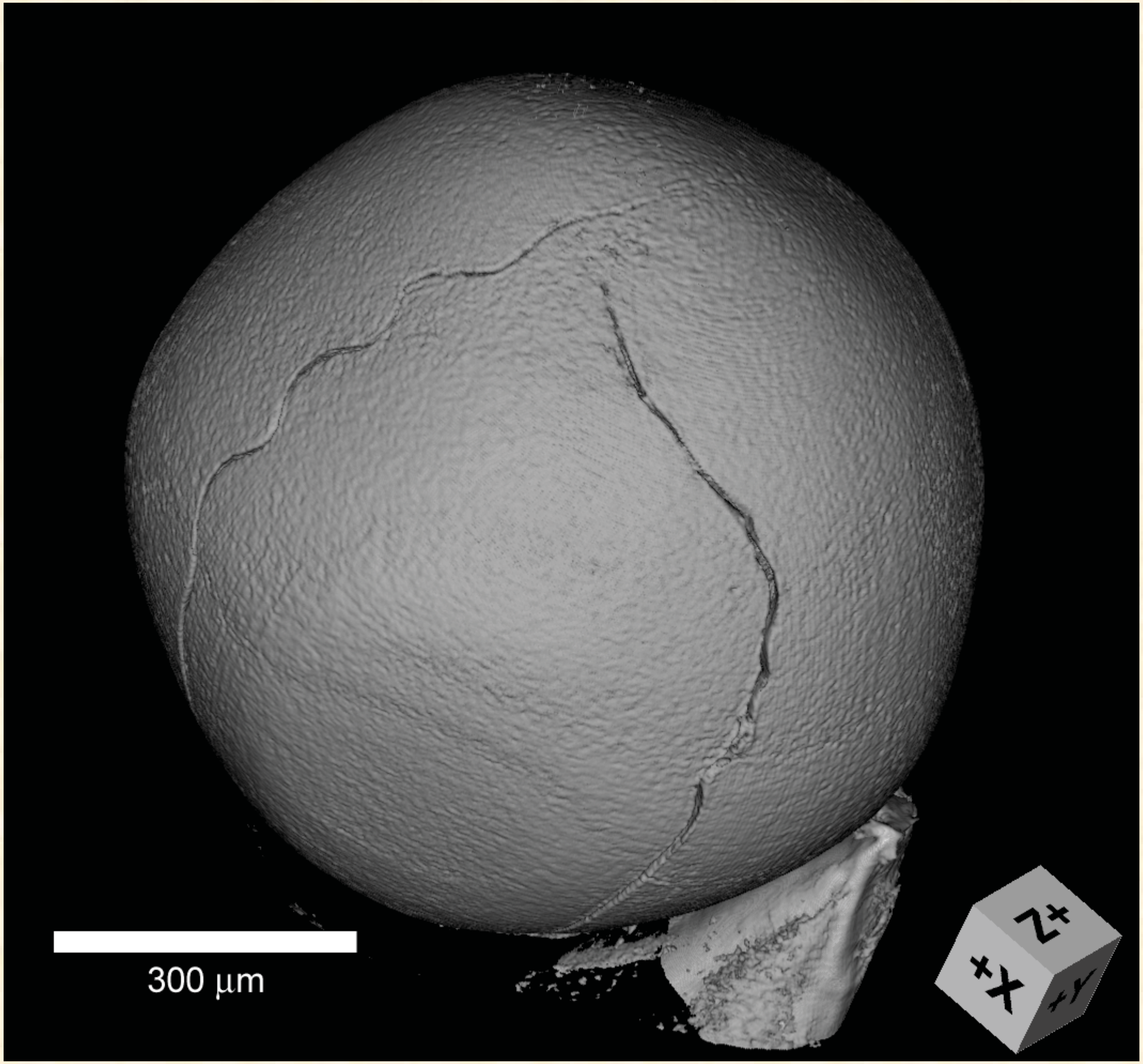
In-depth analysis of defective TRISO particles

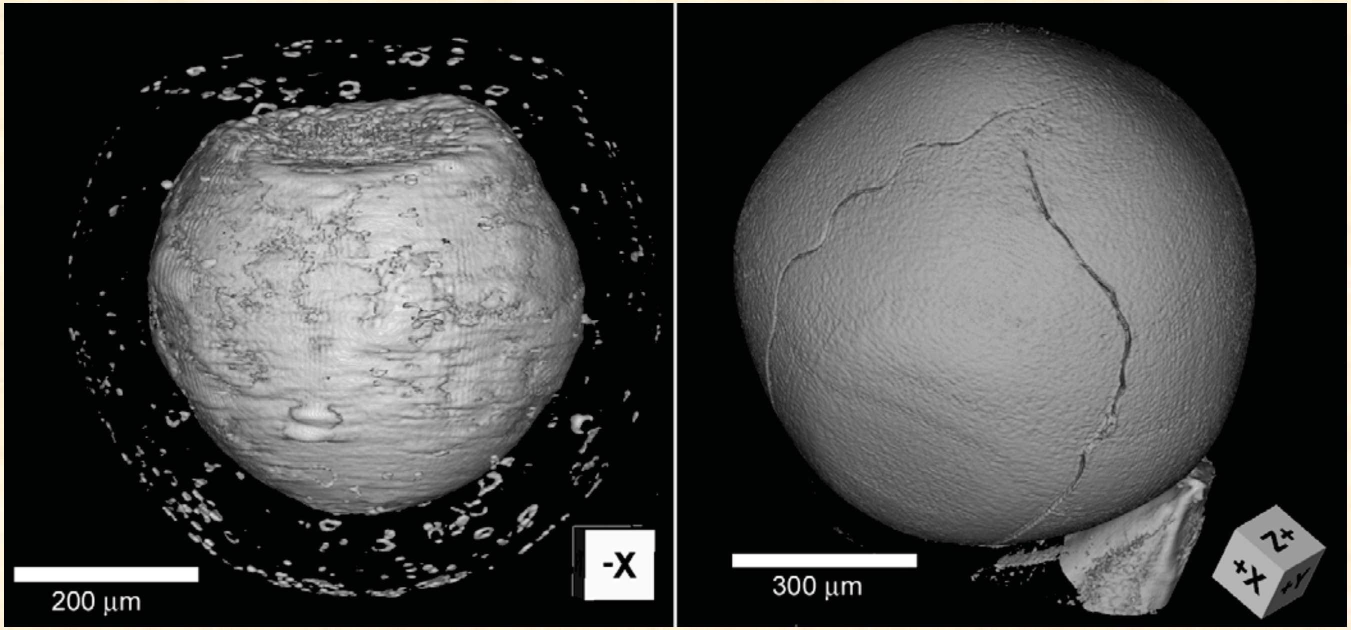


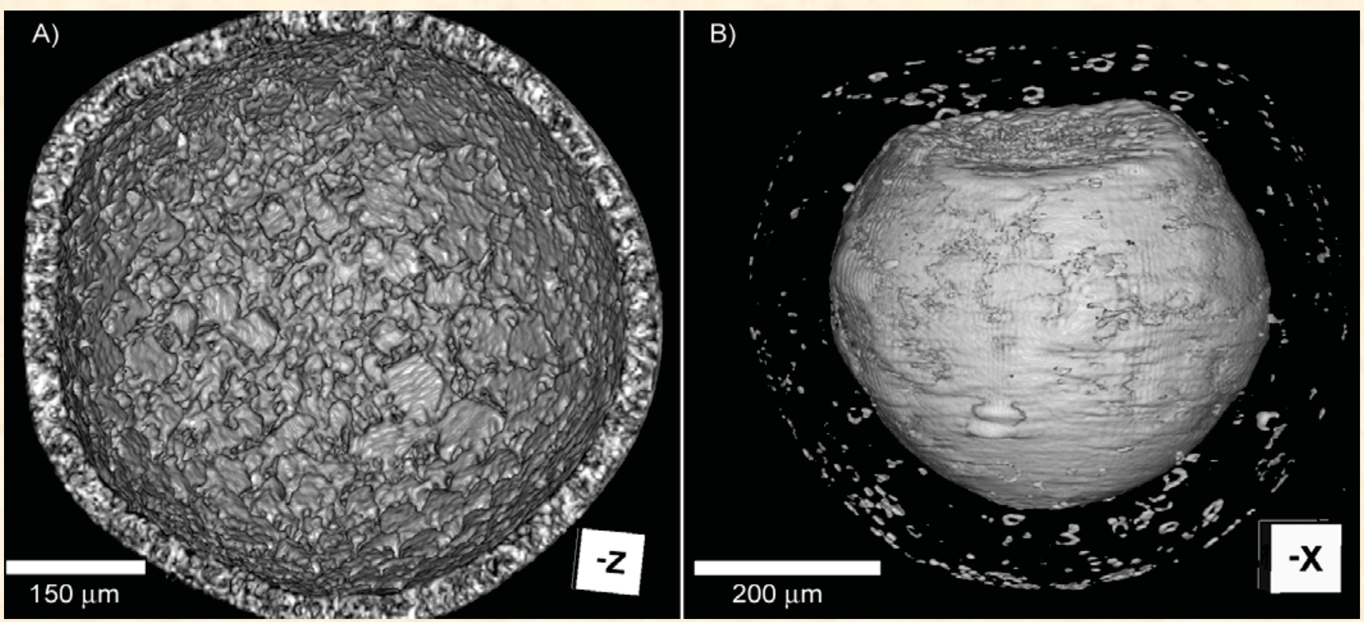
Animation:
looking at 2-D slices going through a TRISO particle

TRISO particle with goldspot which was identified during visual examination of particles with OPyC burned off

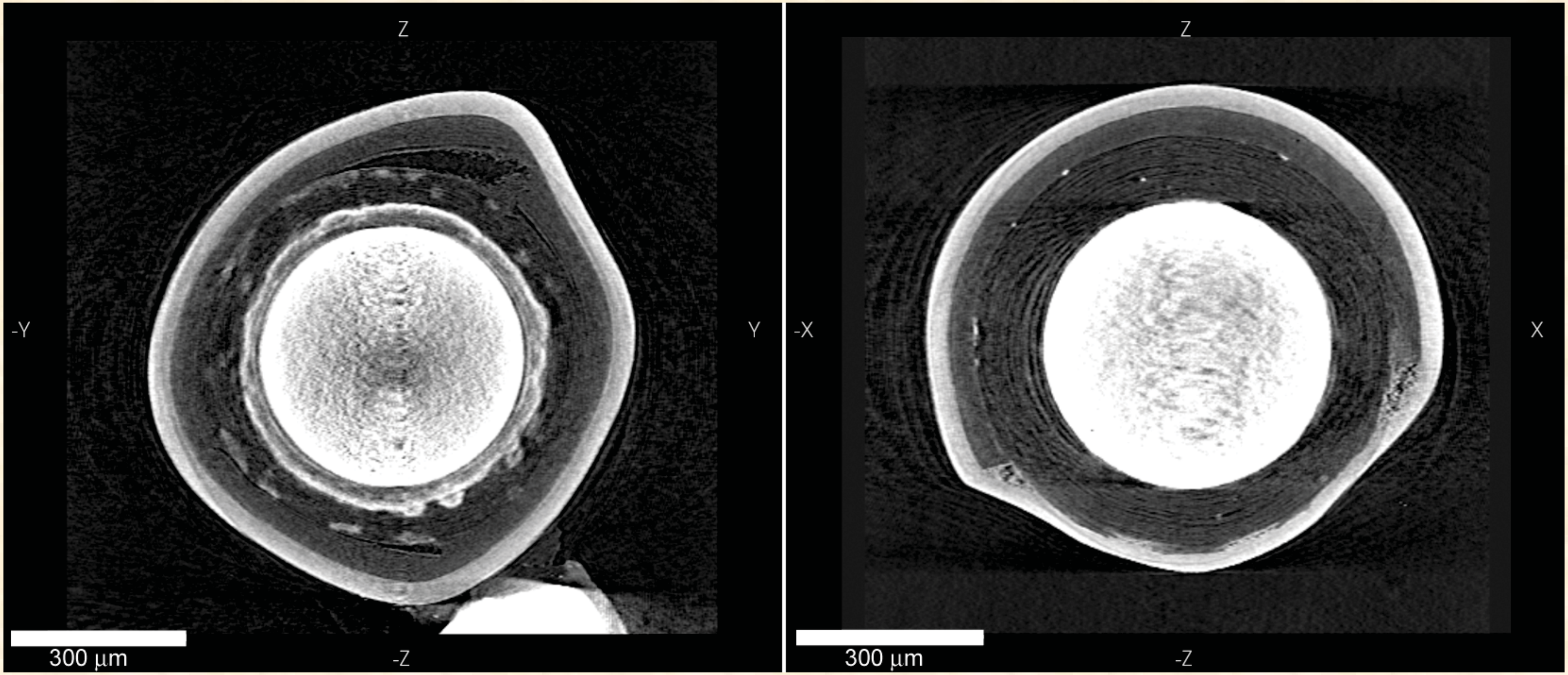
Crack in SiC Layer



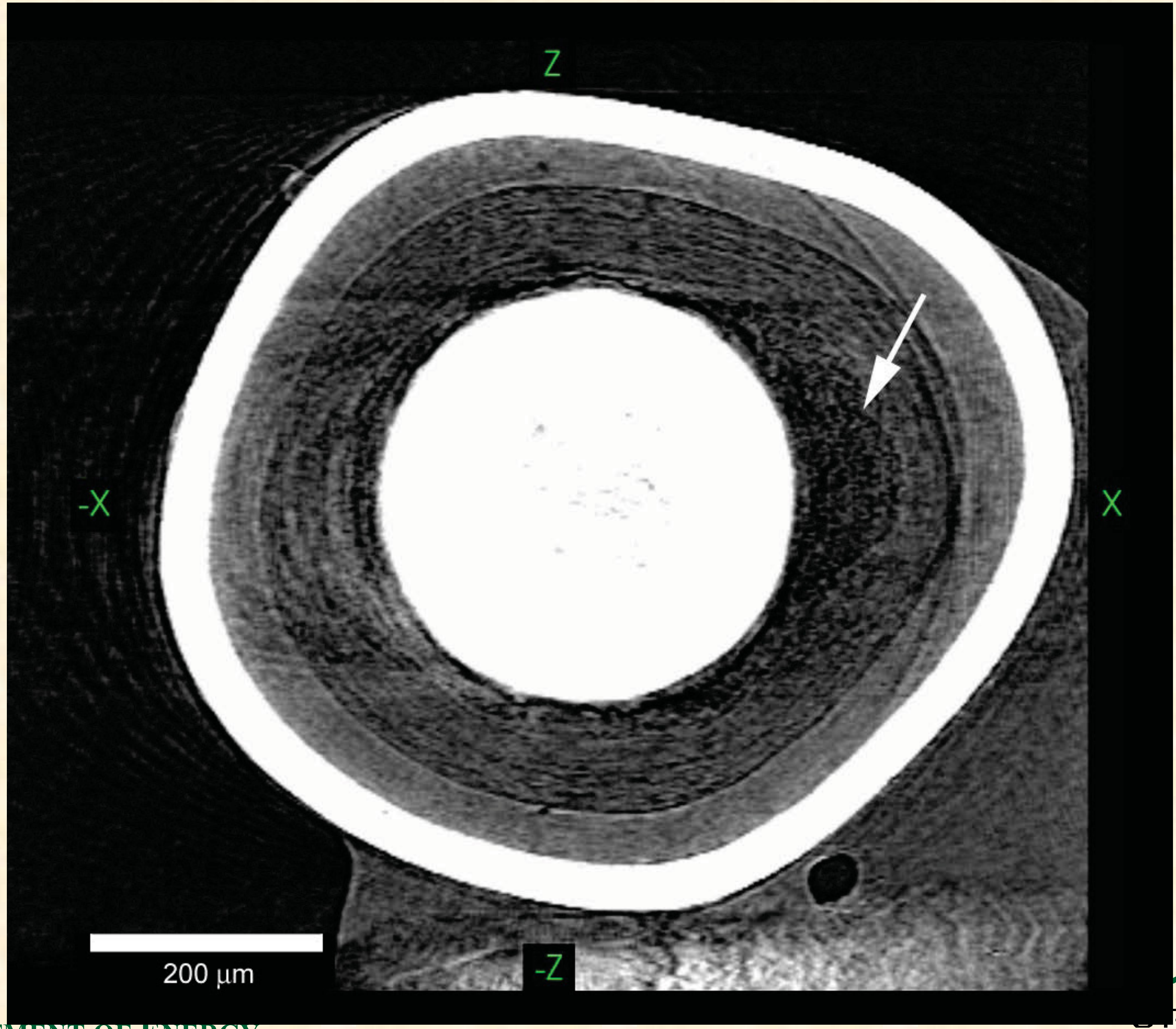

-X
200 µm
300 µm
Z+
+X
+Y


A)
-Z
150 µm
B)
-X
200 µm

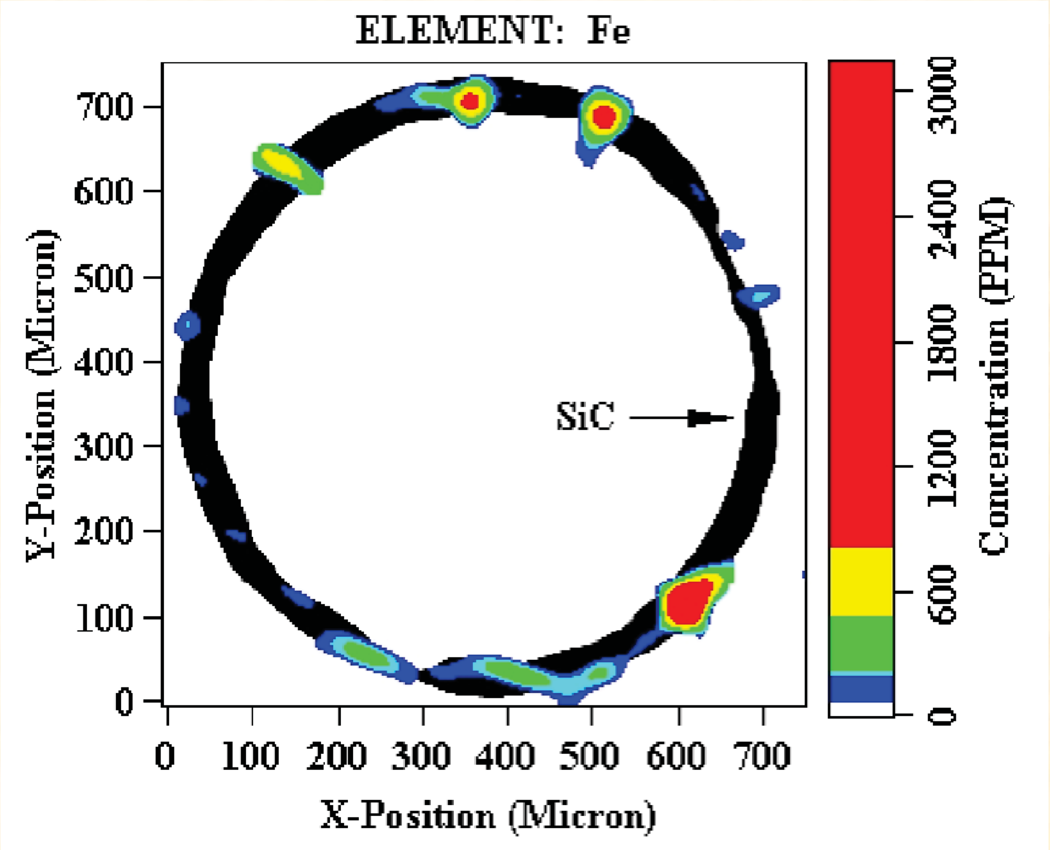
Broken IPyC



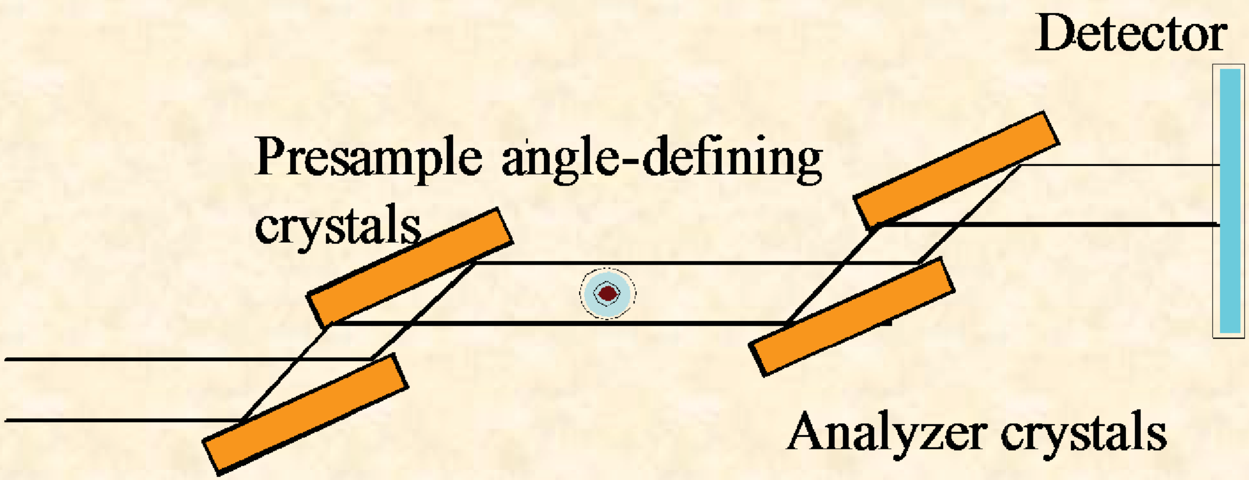
Buffer Suit



Syncrotron Tomography for Elemental Analysis



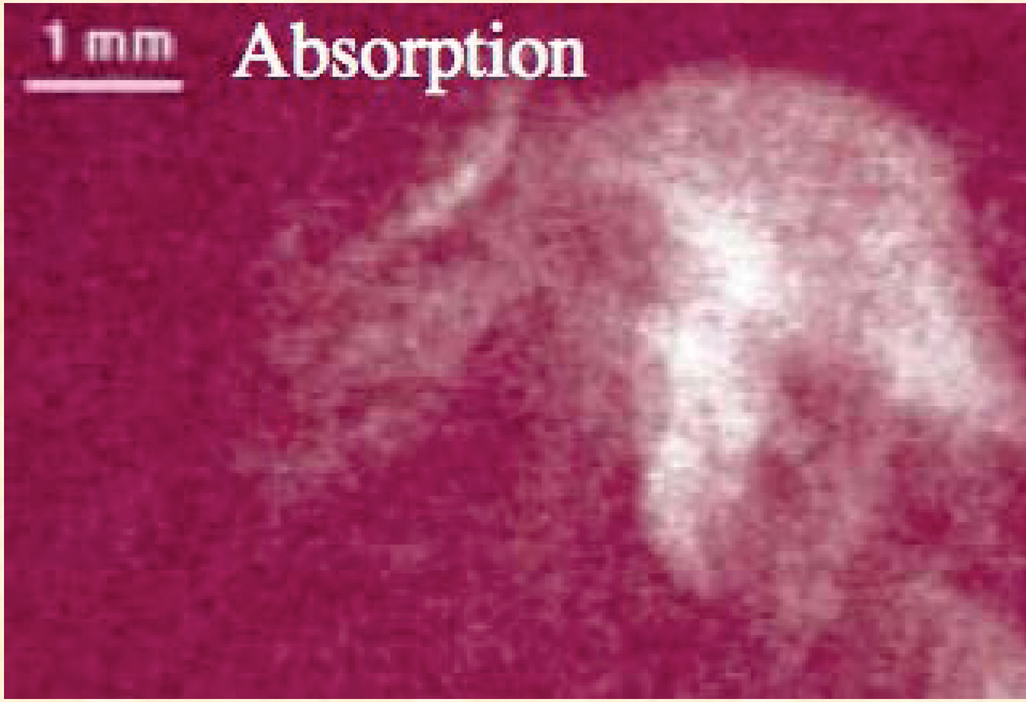
Even better performance possible: *Diffraction contrast* dramatically improves contrast in low Z materials

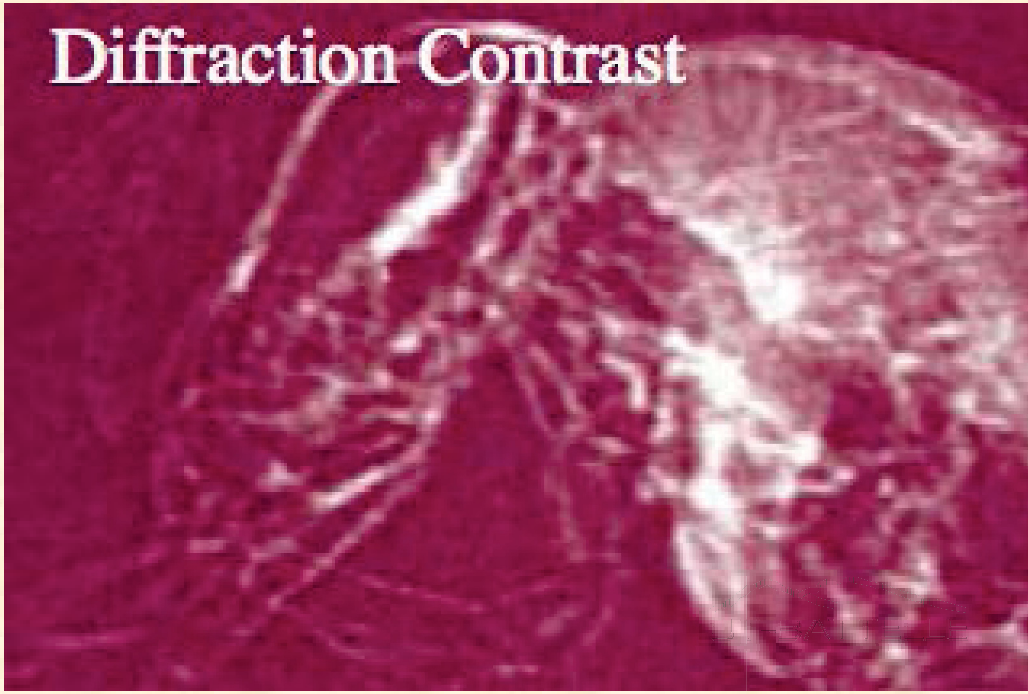


- Light elements like carbon have low x-ray absorption
- Diffraction contrast is sensitive to the refraction of x-rays, a stronger effect for these elements.
- Detector is also located further away from sample (less rad-sensitivity.)
- This technique is being developed at synchrotrons; we will use advanced x-ray optics to implement it using a laboratory x-ray source

Diffraction Contrast Imaging
- ongoing upgrade -

- Orders of magnitude better contrast
- Particularly sensitive to surfaces, cracks, voids, and inclusions
- We will be able to *detect* features which are smaller than the instrumental resolution (e.g. fine cracks)





Concluding Remarks

- Current tomography unit is being applied in the research mode (research on the technique, equipment, and fuel) for TRISO fuels and graphite.

- Current resolution is ~ 1.4 micron. Effort is underway (within a year) for sub-micron resolution and improved contrast. Technique has been demonstrated with 70 nm resolution.

- Current limitation is ~ 100 mR/hr @ one foot. (LAMDA guideline.) Both the newly installed LAMDA FIBs and the Tomography unit are being rad-hardened and having transfer/loading fixtures which will allow higher dose samples.

- This instrument has been applied to metallic samples (steel, tungsten.) Due to higher Z the samples must be significantly smaller. However, this is consistent with fundamental studies for metallic fuels.

Current and Future Micro-analysis Devices in the LECA-STAR Facility

J. LAMONTAGNE

Commissariat à l'Energie Atomique et aux Energies Alternatives
Cadarache Center
Fuel Studies Department
13108 St Paul Lez Durance cedex, France

Some essential questions need R&D activities

A large number of questions, sometimes asked from many years

…including the transmutation targets

- Formation mechanism of the nano-scale bubbles?
- Transition mechanism from nano-scale bubbles to micro-scale bubbles?
- Link between the grain size / release, grain boundaries behavior (irradiation, ramp, heat treatment)?
- Parameters involved in the HBS (High Burnup Structure) formation?
- Gas release during the HBS formation?
- HBS evolution at very high Burnup?
- Existence of Fission Products compounds in intergranular position in the HBS? other phase?
- Precipitation and release gas mechanism during an annealing test? Movement of the bubbles?

… and many more

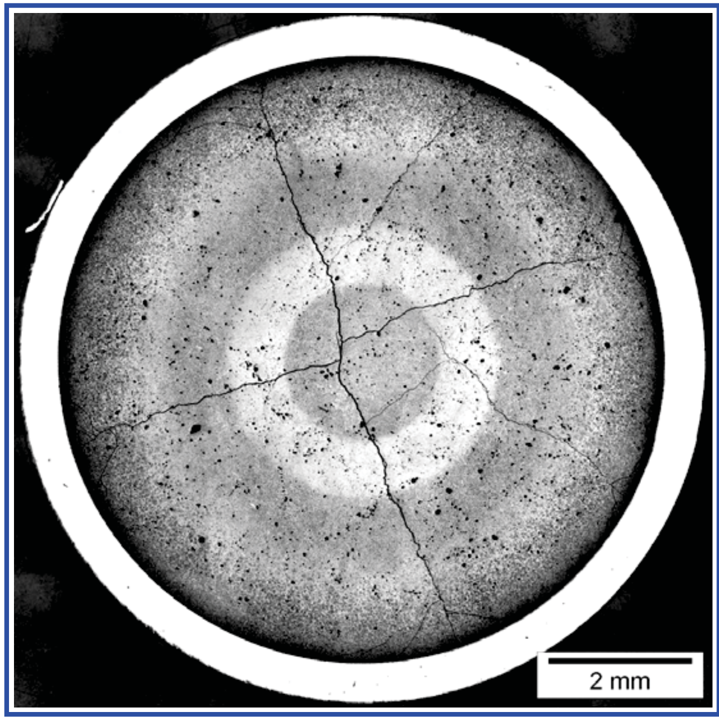
Strategy:

- Mechanical approach
- Physical chemistry approach

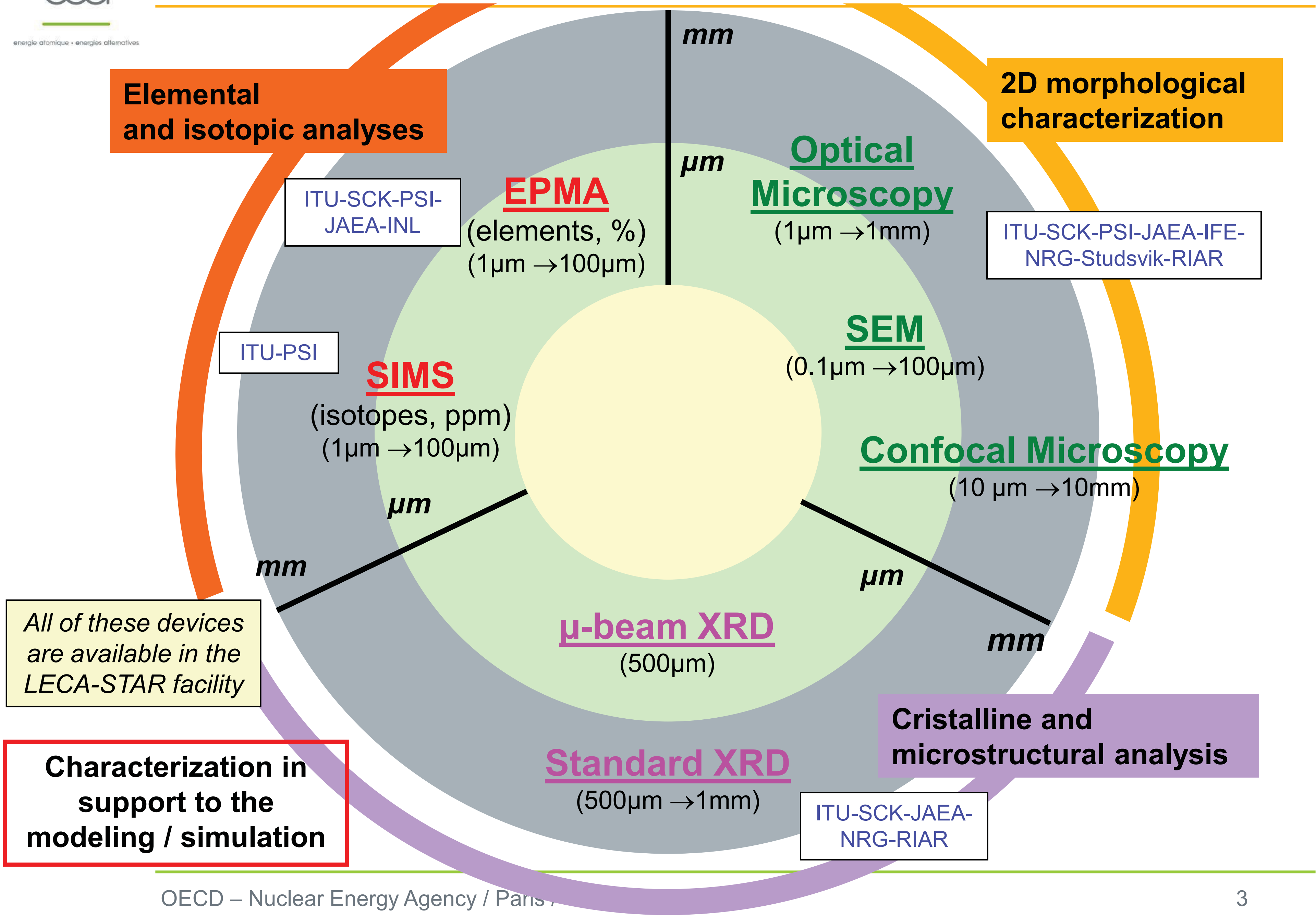
↳ **multi-scale analyses**

Optical Macrography of a fuel radial (BU ~80 GWd/t)

HBS in periphery, precipitation en central area (0R to 0.26R) + ring precipitation around 0.55R



Characterization of the irradiated fuel: from the *mm* to the *µm*



ECRIX-H Irradiation Experiment

Basic framework:

Heterogeneous monorecycling of Minor Actinides (MA) under a locally moderated neutron flux in Na-cooled Fast Reactors (SFR)

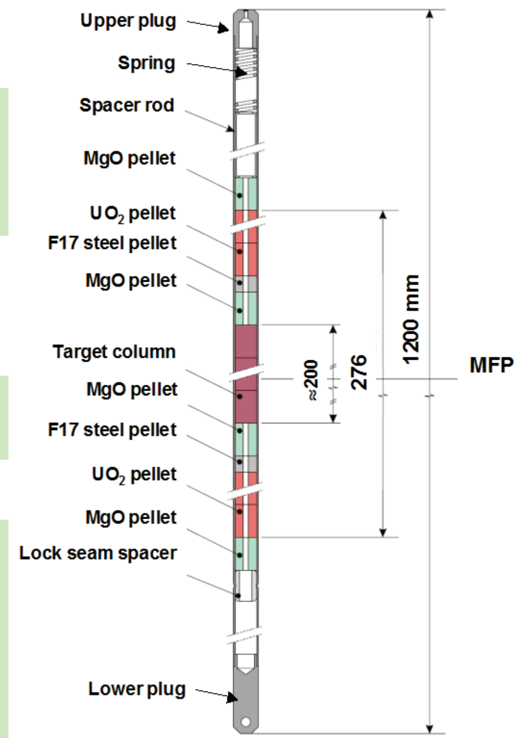
Béjaoui et al, JNM (2011)

Transmutation targets:

$AmO_{1.62}$ microdispersed in MgO with 0.66 g of Am/cm^3

Irradiation in Phenix core during 318 EFPD

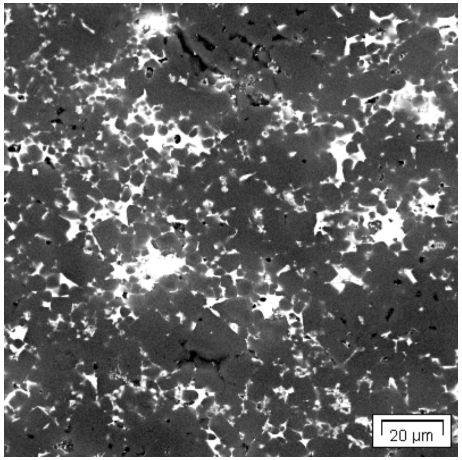
Expected transmutation and fission rates from pre-irradiation simulations: → 92.6 and 33.9 at% (at the maximum flux plane)



PIE Results:

Satisfactory behavior of the MgO-AmO$_{1.62}$ targets (moderate swelling) from a <u>macroscopic</u> point of view

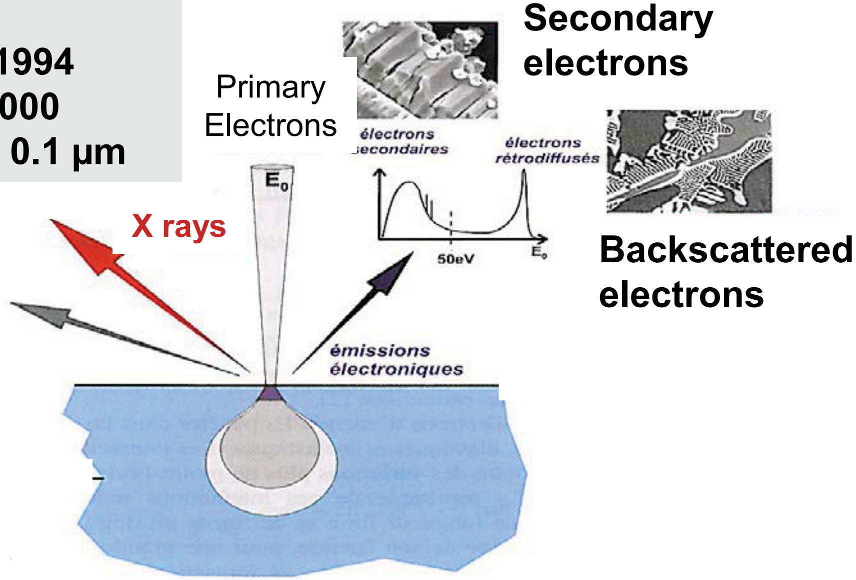
Lamontagne et al, JNM (2011)



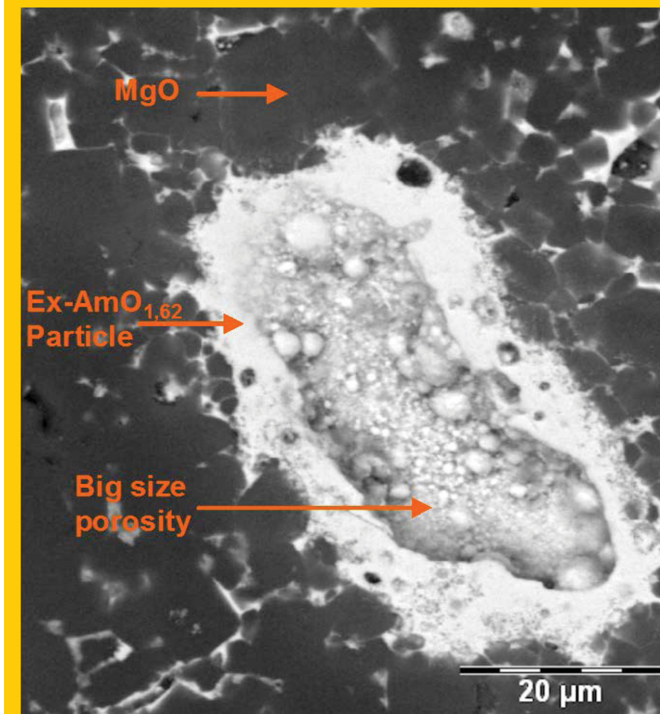
Microscopic point of view?

SEM (Scanning Electron Microscope): morphological analyses

SEM XL 30 (FEI)
In operation since 1994
Magnification: x 10000
Lateral Resolution: 0.1 µm

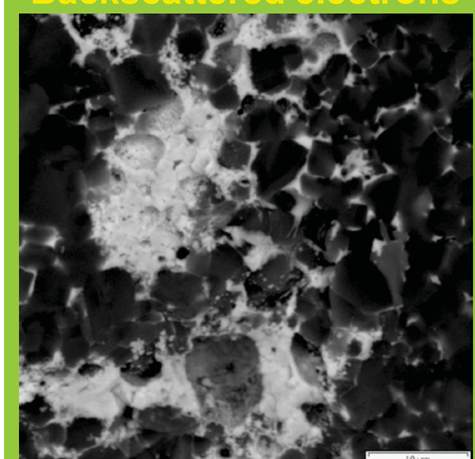


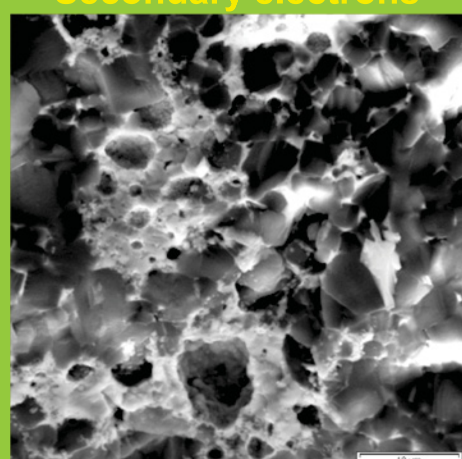
Polished sample



Fractography

Backscattered electrons | **Secondary electrons**





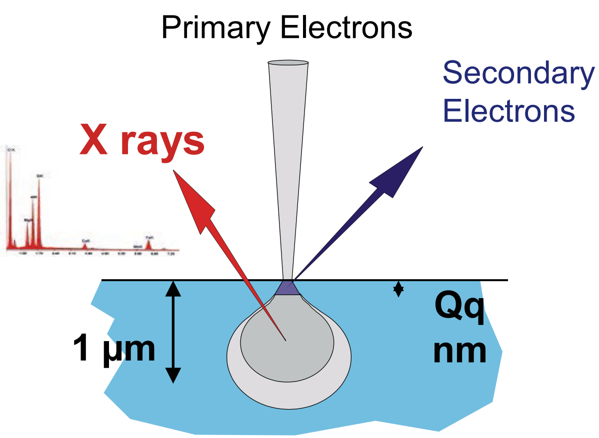
Characterization along a radius:

- Porosity (in, out the ex-AmO particle, interface)
- HBS
- MgO grains fractures

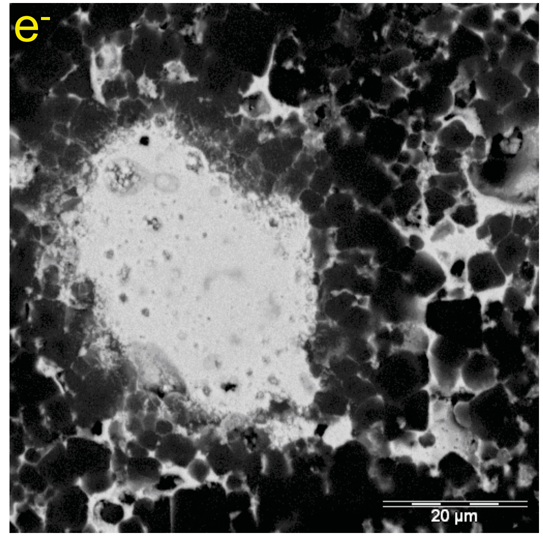
➢ *Nanoscale bubbles and their formation mechanism?*
➢ *Transition mechanism from nano-scale bubbles to micro-scale bubbles?*

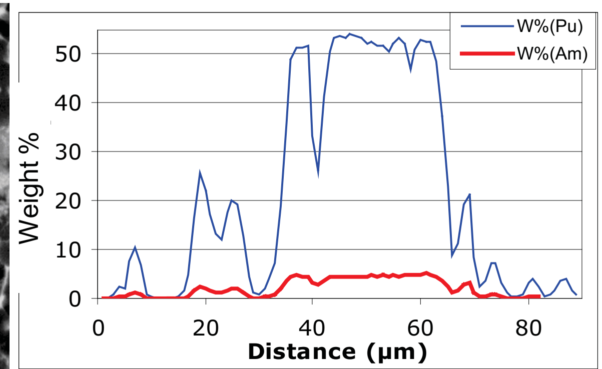
EPMA (Electron Probe Micro-Analyser): chemical analyses

Shielded EPMA CAMECA SX100R
In operation since December 2009
Detection: < 1 %
Lateral Resolution < 1 µm

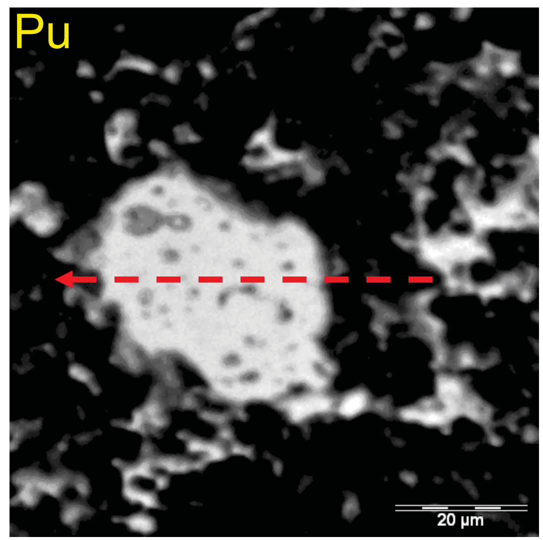


Distribution and content:
Fission Products, actinides …

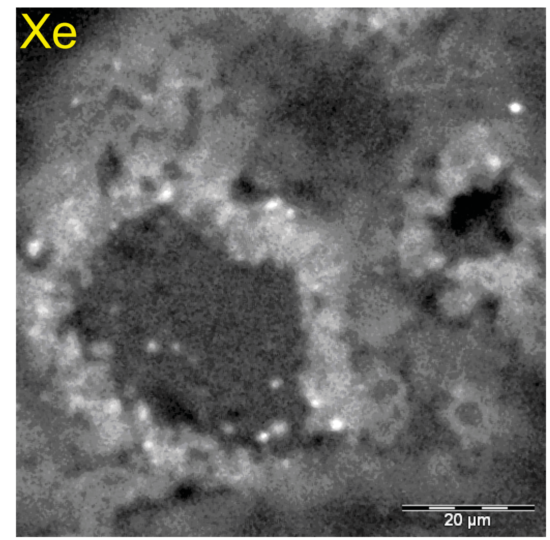




Transmutation rate: ~95%



Pu formation in the ex-AmOx particle

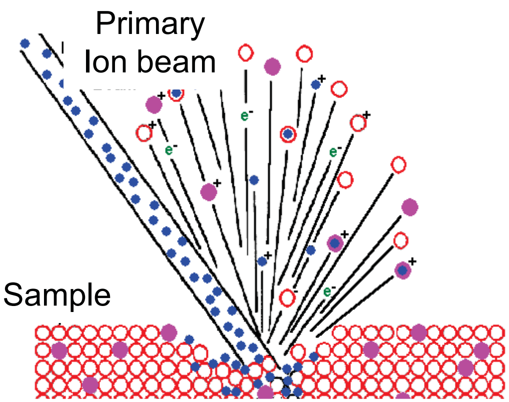


Xe (+others FPs) departure from particle

- *Mechanism of the FPs Movement?*
- *Nanoscale position of the Fission Products compounds?*

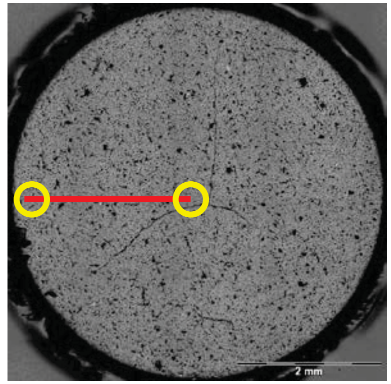
SIMS (Secondary Ion Mass Spectrometry): isotopic analyses

Shielded SIMS CAMECA 6F
In operation since 1998
(2002 for irradiated fuel)

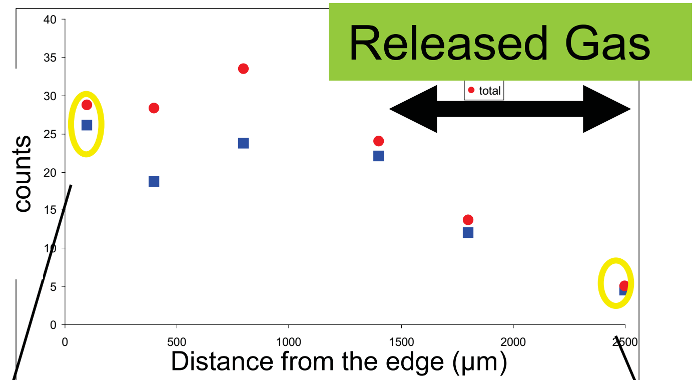


- Isotopic analyses
- Trace elements (ppb)
- Gas inventory

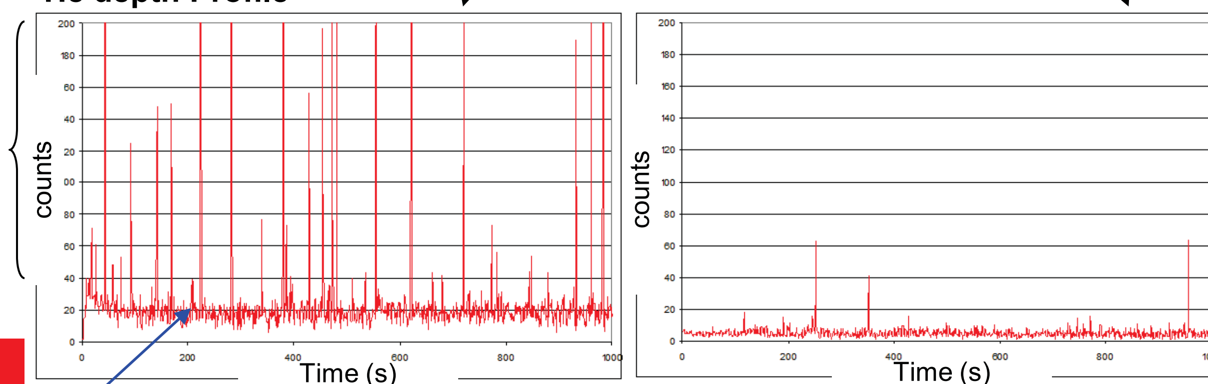
Optical macrography of a radial cut



He radial Profile



He depth Profile



Peaks
(bubbles full of gas)

Baseline
(~Gas dissolved in the matrix)

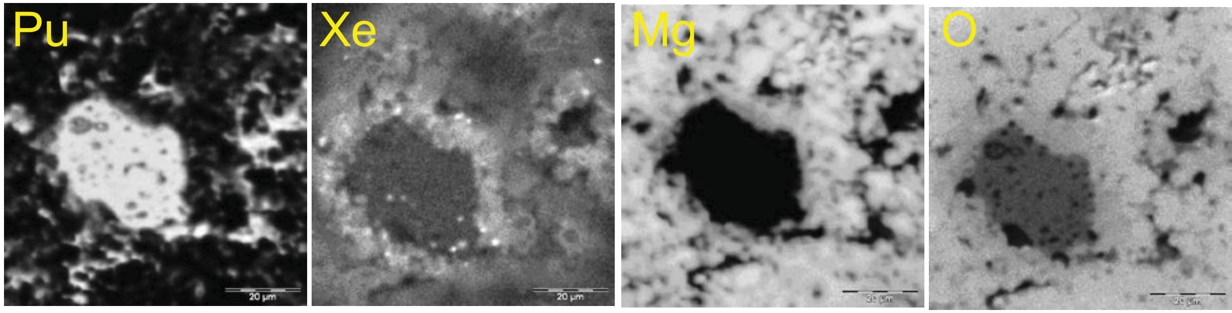
➢ *Mechanism of the gas release?*

XRD (X-Ray Diffraction): cristalline analyses

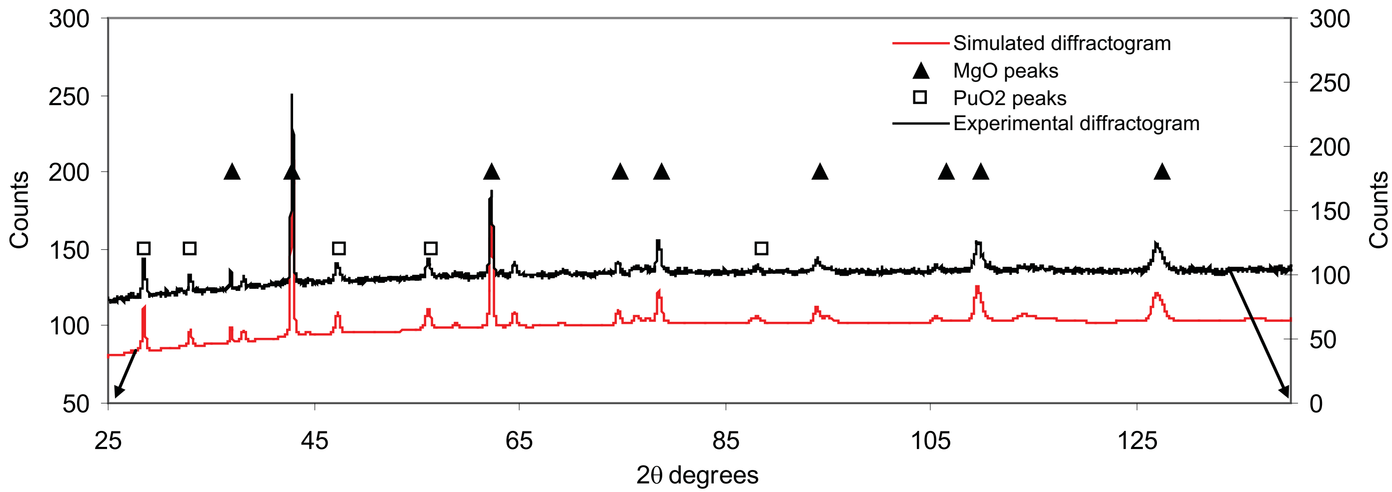
General Electric
Micro-beam Module (500 µm)
In operation since December 2008

- **Phases** (identification, phase %, cristalline evolution…)
- **µ-distortion and strain**

EPMA mappings



Quantitative Phases Analyse



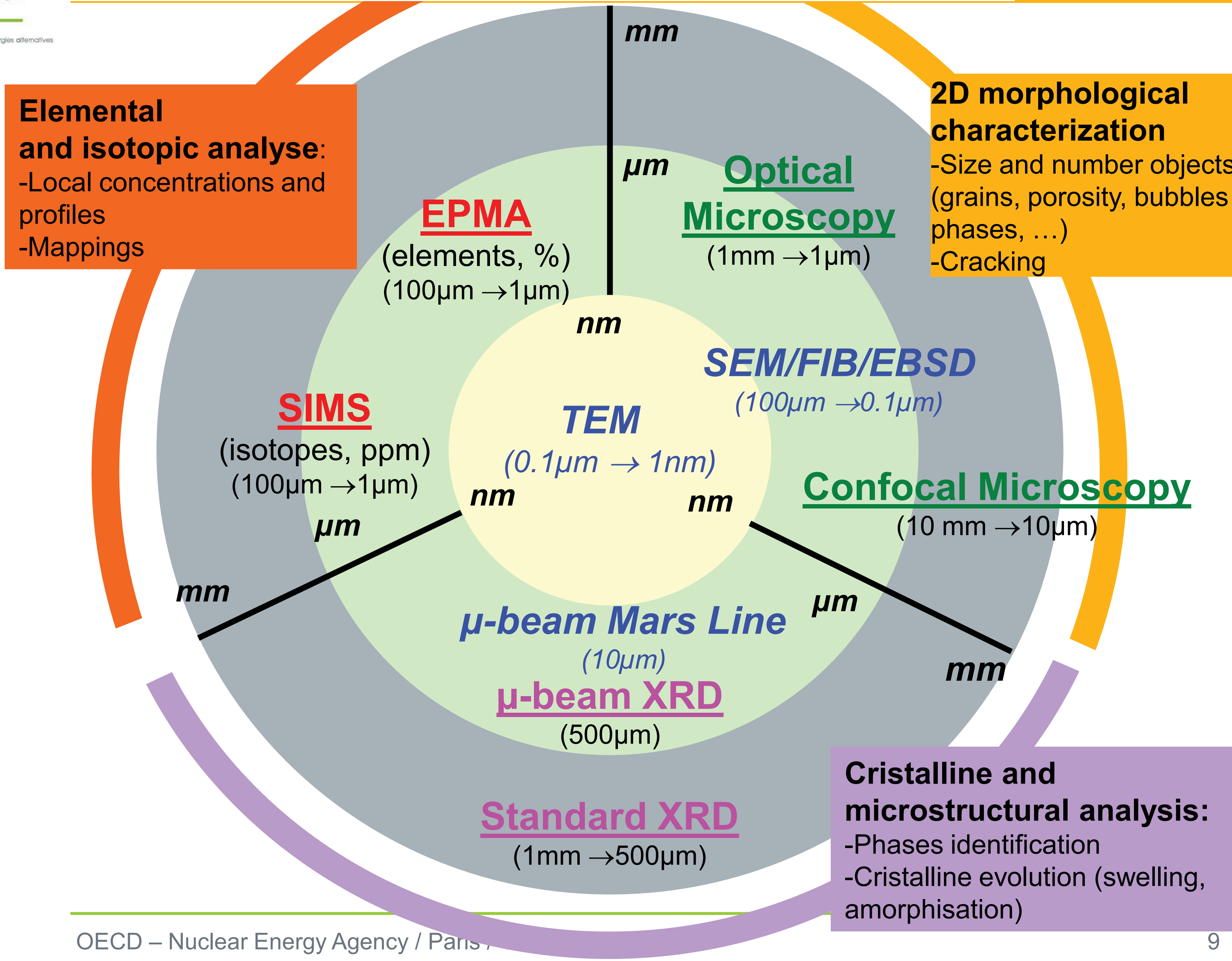
MgO = 94.4 w%

PuO_2 = 5.6 w%

Consistent with the EPMA analyses and a high transmutation rate

- ➢ *Which structure for the few Am remaining (detected by EPMA but not by XRD)?*
- ➢ *Which strain in the MgO grain at the periphery of the particle (FPs implantation effect)?*

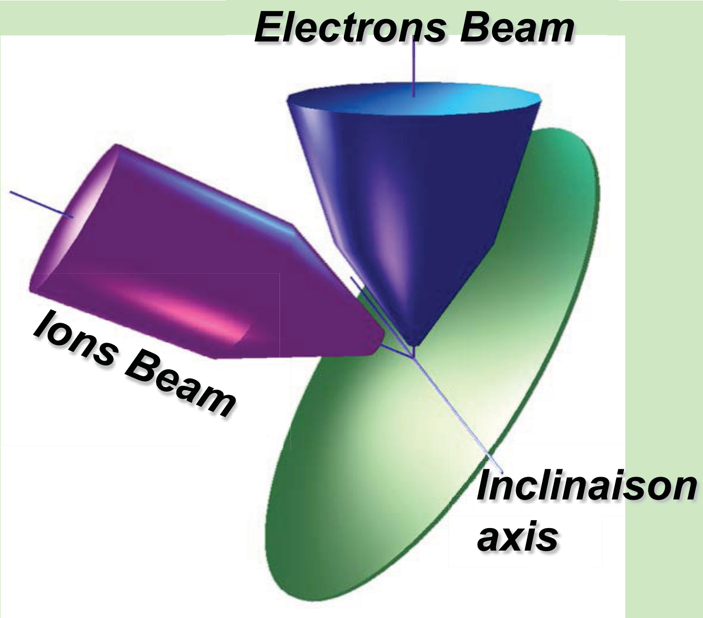
Perspectives: from the millimeter to the nanometer



SEM-FIB (Focused Ion Beam)

Principle

- **SEM with a ionic beam**
- Electronic column + ionic column
- The 2 beams are focused on the same sample position



Project status:

- ➢ **Installation in the hotcell in place of the current SEM by 2014**
- ➢ **Commercial process ongoing**

Progress for the irradiated fuel:

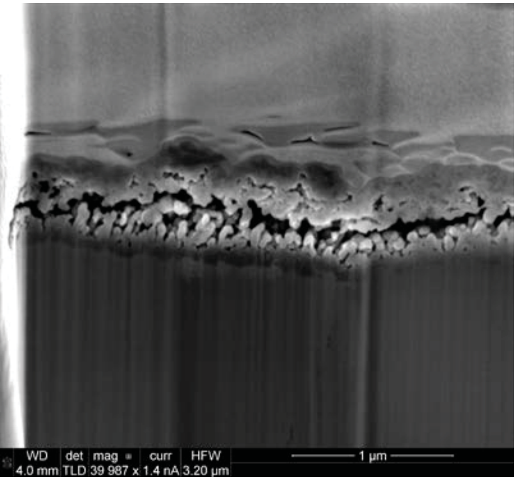
• 3D imaging of the microstructure: gas behavior (tunneling, bubbles location and interconnection), cracking

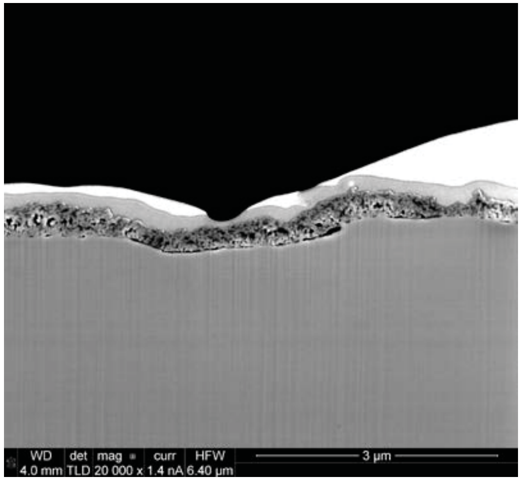
• Preparation of thin samples

SEM-FIB: contribution #1

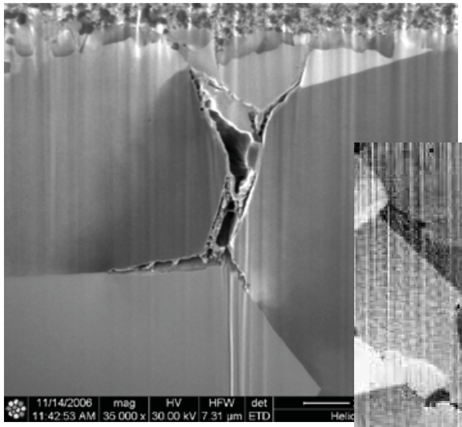
Electronic imaging during ionic sputtering

Corrosion layer

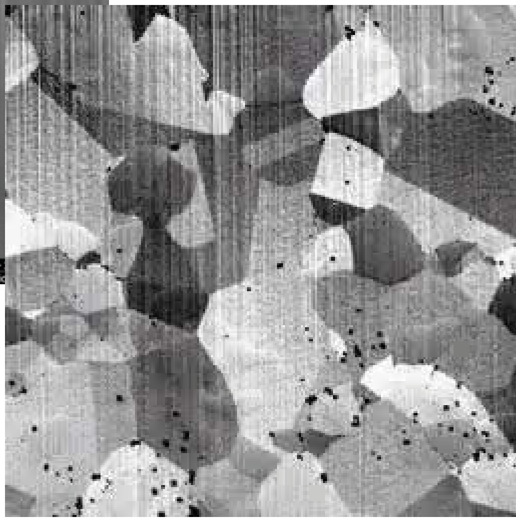




Crack



Grains and bubbles location



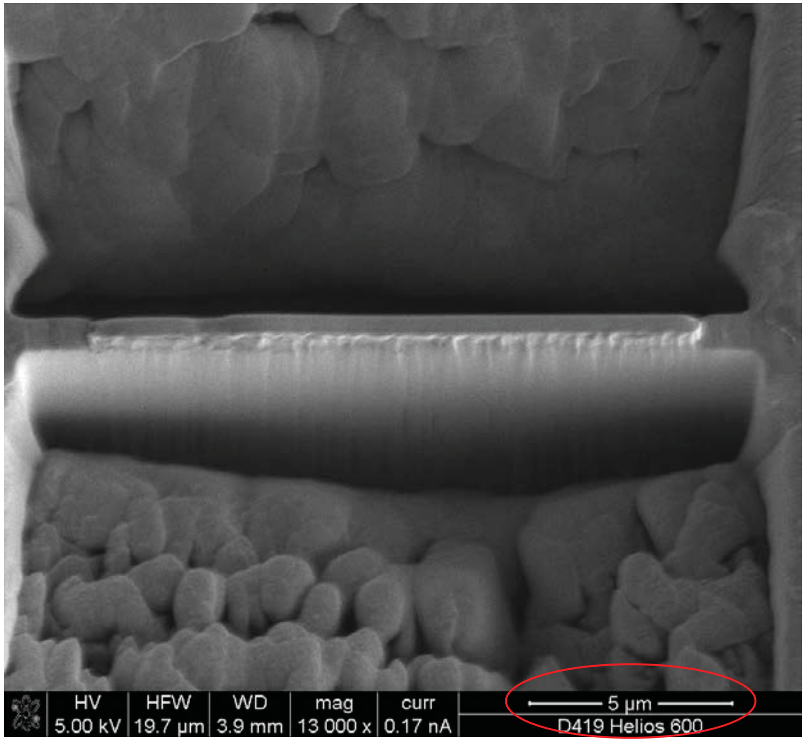
Progress for the irradiated fuel:

- **Study of a delicate sample without mechanical polishing (ex: porous sample)**
  - **→ no damage**
  - **→ for EBSD analyse**
- **Cracks: study of the cracks initiation and the propagation**
- **Study of the thin porosity distinguishing inter and intragranular, interconnection**

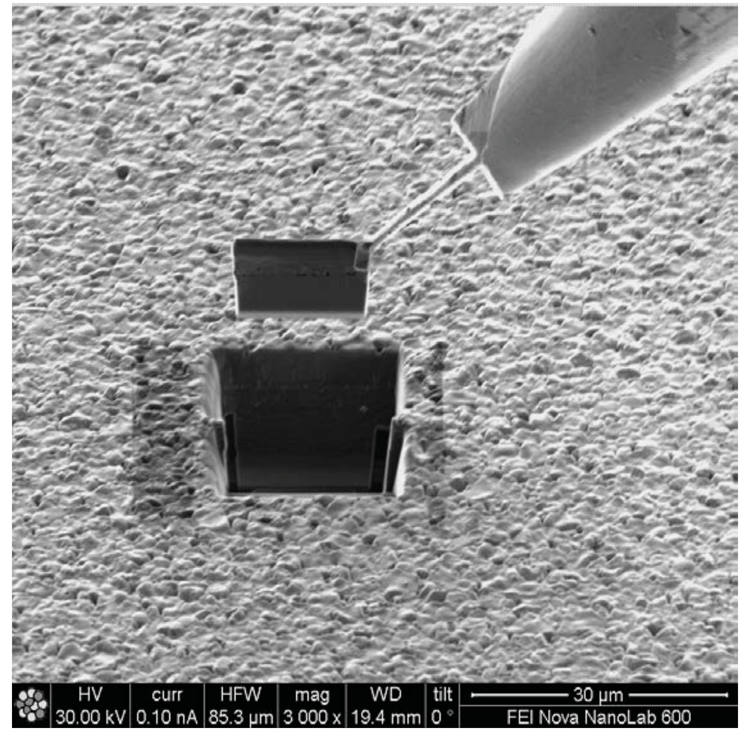
SEM-FIB: contribution #2

Preparation of small and/or thin samples

Cutting of the sample



Sample extraction



Progress for the irradiated fuel:

→ Irradiated sample preparation for the TEM

→ Irradiated sample preparation for the Mars Line

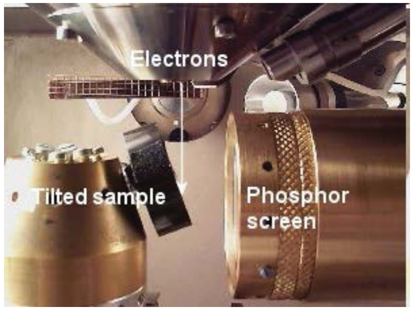
→ Choice of the sample location (AmOx particle, MgO, Interface)

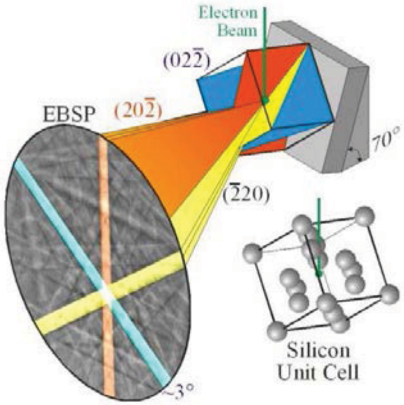
→ Reduction of the radioactive background for the current microanalyse devices (XRD, SEM, EPMA)

EBSD (Electron BackScattered Diffraction)

Principle

Associated with the SEM-FIB





- Interaction between a crystal and the primary electrons
  - Diffraction of the primary electrons by the cristalline plans
  - Acquisition of a diffraction pattern
  - The cristalline structure is obtained on a grain scale
  - High quality of sample preparation is needed → SEM-FIB

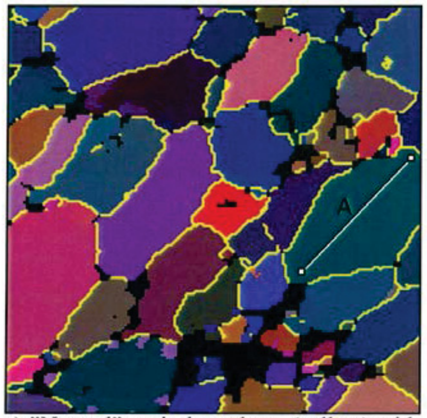
Project status:

- **Installation associated with the SEM-FIB**
- **Installation in the hotcell in place of the current SEM by 2014**
- **Commercial process ongoing**

EBSD: example of contribution

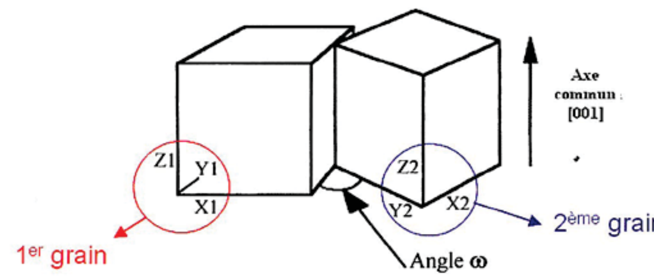
Displaying of the grain boundary by SEM/FIB/EBSD

→ Correlation between the grain restructuration and their cristalline orientation



c) "Normal" grain boundary (yellow) with >10° mismatch between two adjacent measurement points. A 6° orientation mismatch between two sides of the same grain is indicated at "A".

d) "Normal" grain boundaries (yellow) and "Sub-grain" boundaries (white) with a mismatch between adjacent measurements in the range 1 and 10°.



Addition of EBSP equipment to the Studsvik HCL SEM – S. Bengtsonn

Meeting 5 and 6 June 1997 Studsvik

Progress for the irradiated fuel:

❖ Phase cristalline study at the grain scale

❖ Extraction of the grain boundaries (→ Porosity location) and the sub-grain (→ restructuration in HBS)

❖ To correlate the cracks propagation and/or the grain restructuration with the cristalline orientation

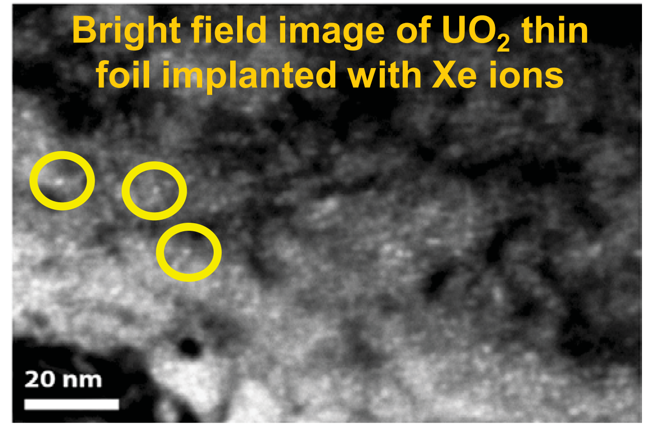
❖ Studying the sub-grain strains during a mechanical test or during the irradiation of the material

TEM (Transmission Electronic Microscope)

At the nano-scale, the TEM allows to…

- Study the **microstructure**
  → *imaging, until to the atomic columns*
- Perform the cristalline study
- Determine the elemental **composition**,
  → *From heavy (EDS) to light elements*
- Obtain some information on the atom **environnement**

C. Sabathier



Xe aggregate size ~ 2 nm

These investigations are performed at the CEA Fuel Studies Department on ion irradiated UO_2 samples
→ Objective: perform the same investigations on irradiated fuels in reactor (all fuels)

Characterisation in support to the modelling / simulation

⇨ an essential device for the irradiated fuel studies

How to perform these PIE:

- ➢ Collaboration with ITU
- ➢ LECA-STAR facility: Project under discussion in the framework of a French invitation to tender for the excellence equipments / installation by the end of 2015

MARS* : A multipurpose beamline for the characterization of radioactive materials

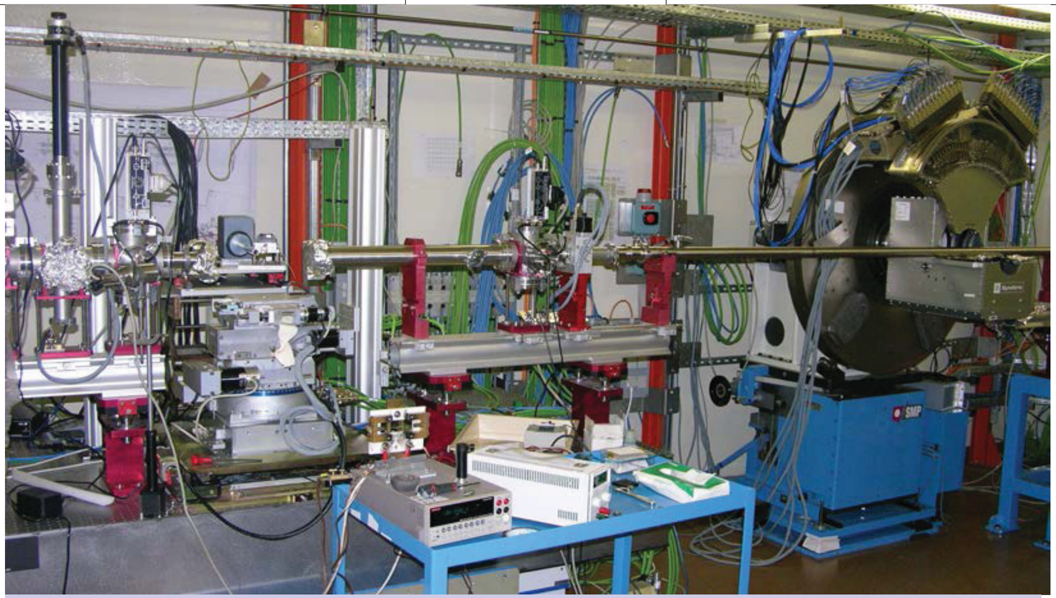
Location: SOLEIL Synchrotron, Saclay, France

*** MAtière Radioactive à SOLEIL or Multi Analyses on Radioactive Samples**

2 experimental stations : 3.5-36 keV

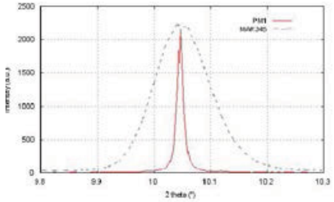
Standard Absorption station

High-resolution XRD



Commissioning on irradiated fuel by the end of 2012

Resolution on LaB6



➔ 0.01°

Further on ECRIX-H experiment

- minor phase detection
- Pu, Am local environment
- (Pu, Am)$O_{(2 \pm x)}$ structure determination
- μ-strain in fissile clusters
- ….

Synchrotron radiation ⇔ XRD station

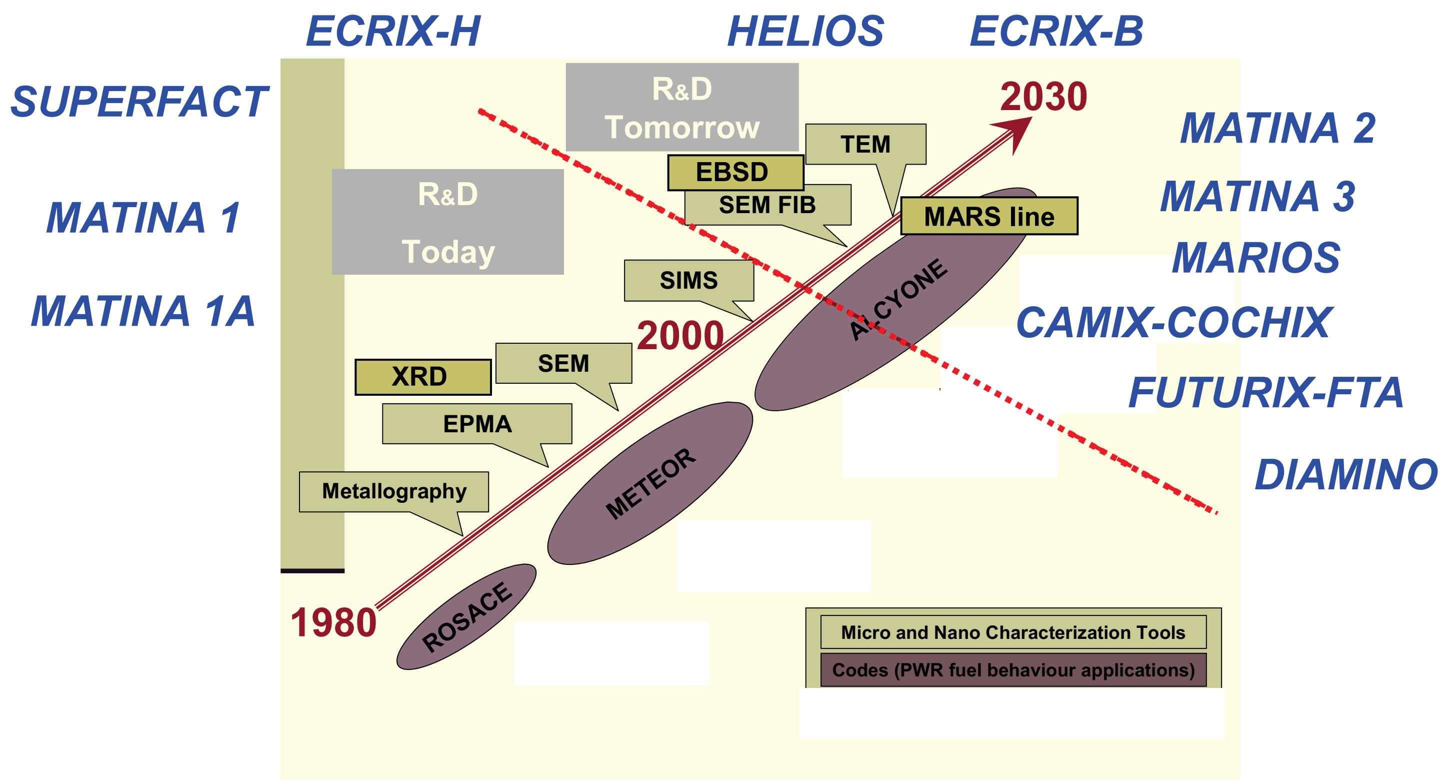
High resolution goniometer + synchrotron flux (10^8 compare to hotlab XRD device)

Conclusions

For the nuclear fuel studies, including transmutation studies, new micro-analysis devices are needed:

❖ **To improve the comprehension of irradiated fuel behavior (including the transmutation),**

❖ **To improve the necessary data for the modeling and simulation**

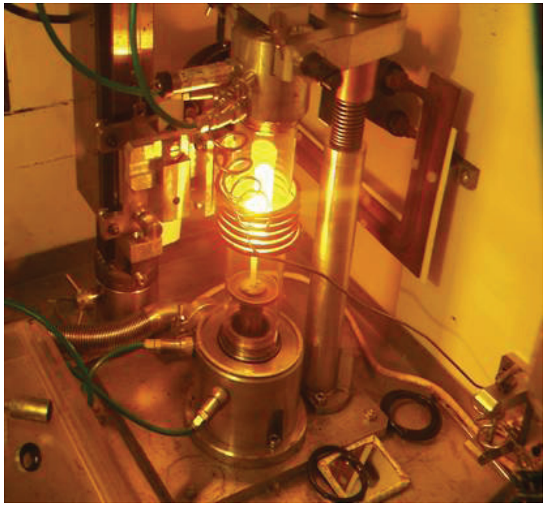


Post Irradiation Examinations LECA-STAR Facility

François SUDREAU

Fuel behaviour characterization and analysis service

MERARG 2
Thermal tests with fission gas release monitoring



- Main characteristics
  - *Sample : from ~100 mg to 10 g*
  - *Induction furnace*
    - Max. temperature : 2800°C
    - Max. temperature ramps : higher than 200°C/sec
    - Atmospheres : He, Ar, air
    - Room pressure
    - Sample temperature measured by 1 thermocouple and 1 pyrometer
  - *Fission gas release kinetics measurements*
    - On-line gamma spectrometry : ^{85}Kr
  - *Post test analysis of released gas (stored in capacities) : gaseous chromatography or mass spectrometry*

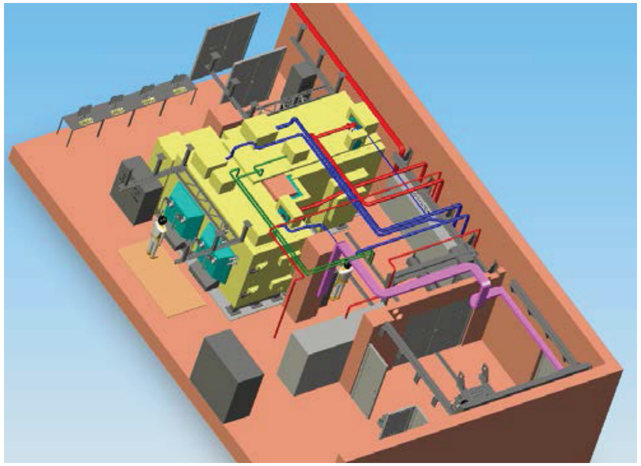
VERDON :
Facility devoted to FP release from irradiated fuels studies

Two adjoining hot cells
in STAR
(CEA Cadarache)

Cell 4:
Storage
Sample preparation
Gamma scanning bench

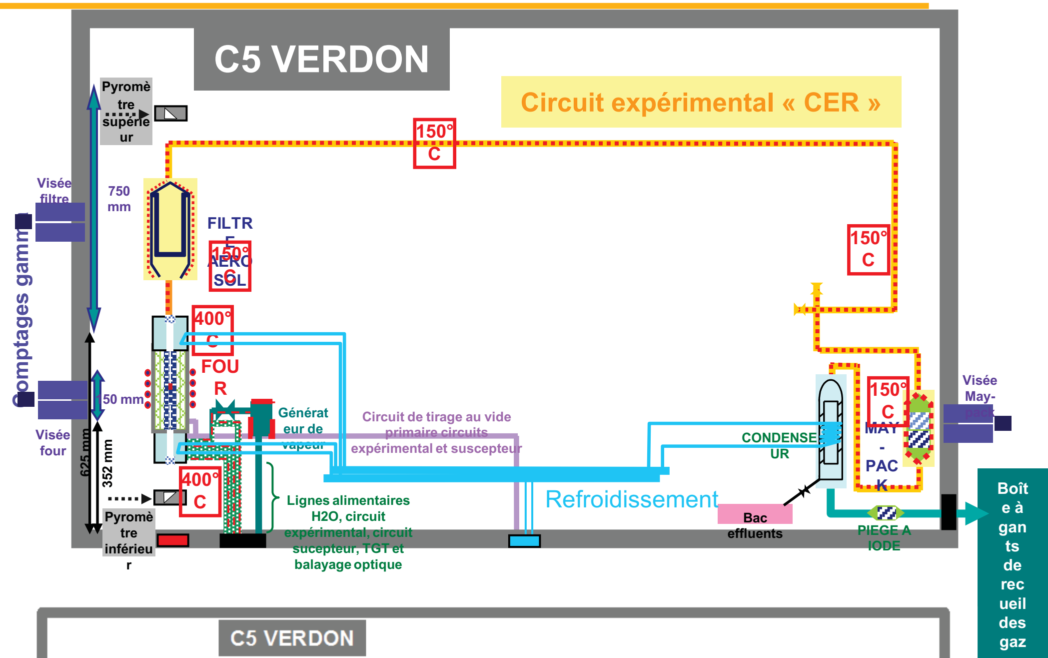
Cell 5:

VERDON loop

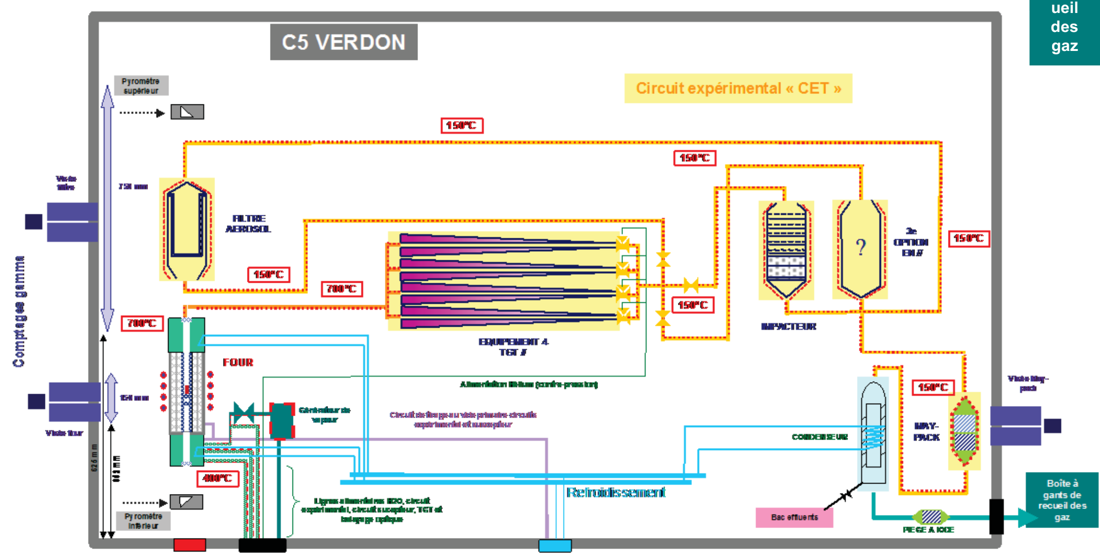


VERDON : 2 loops configurations

- VERDON loop, "Release" configuration (CER)



- VERDON loop, "Transport" configuration (CET)

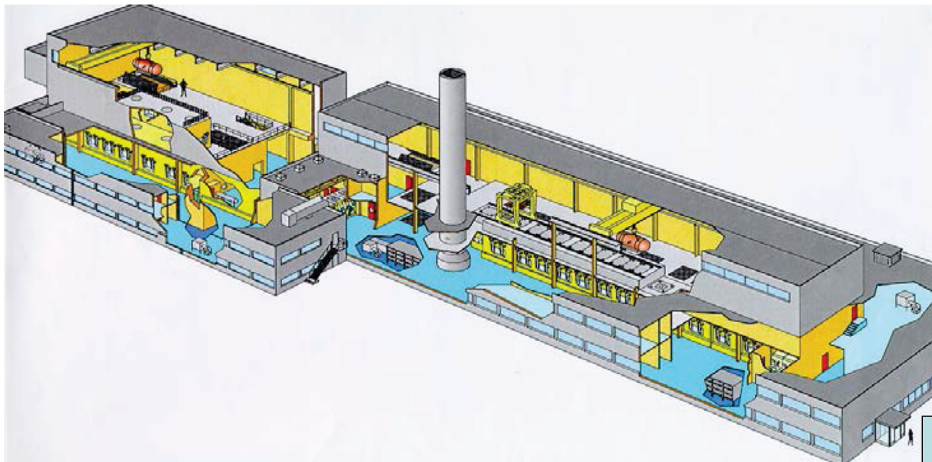


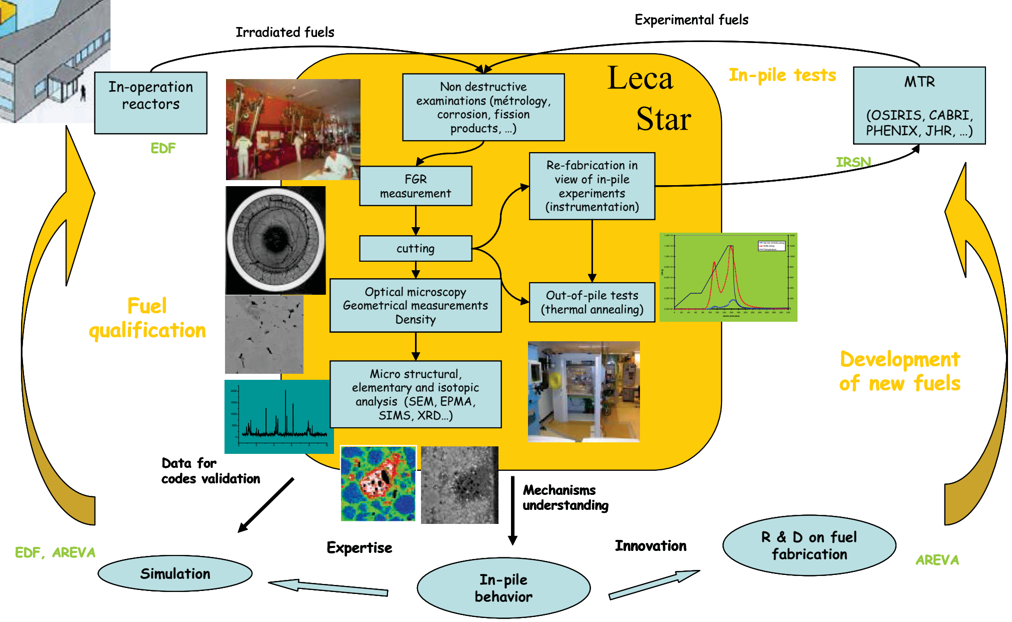
VERDON : Test conditions and on-line measurements

- Fuel sample
  - *"pellet scale" for FP release; 10 cm fuel long for studying the coupling between fuel degradation and FP release*
  - *Reirradiation in MTR reactor for short half life FP*
- Tests conditions
  - *Temperature up to fuel delocation or slightly more: inductive heating*
  - *Tests performed under variable atmosphere: helium, hydrogen, steam, air*
- On line FP measurements
  - *3 on line gamma spectrometers in sight of: fuel, filter, maypack*
- FP measurements before and post tests
  - ***1 gamma spectrometer "multi-samples"***
    - FP balance before and after the test : FP deposit quantification along the loop downstream the furnace
  - *1* ***glove box for fission gas total balance***
    - 1 chromatograph coupled with mass spectrometer: quantification of released stable gas

LECA-STAR

A complete set of devices to study fuel behavior under various conditions





Joint Research Centre (JRC)

Application of X-ray absorption to density measurements

D. Papaioannou (presented by J. Somers)

ITU - Institute for Transuranium Elements

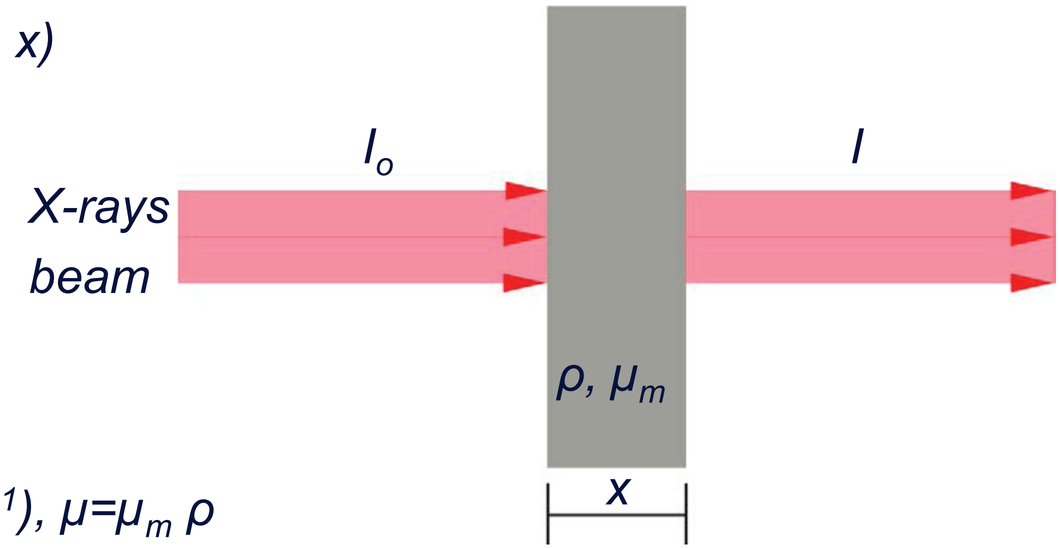
Karlsruhe - Germany

http://itu.jrc.ec.europa.eu/

http://www.jrc.ec.europa.eu/

Attenuation law

$I = I_o \exp(-\mu_m \rho x)$ or $\rho = \ln(I_o/I) / (\mu_m x)$



I transmitted intensity
I_o incident intensity
μ_m mass absorption coefficient (cm^2/g)
ρ density
x sample thickness

Note: μ linear absorption coefficient (cm^{-1}), $\mu=\mu_m \rho$
μ depends on the wavelength, λ
Mixtures and compounds: $\mu_m = \Sigma (w_i \mu_{mi})$,
w_i is the weight fraction of the i^{th} atomic constituent

Assumption: μ, μ_m is independent on x, hence homogeneity

Literature source

"Density measurements of liquid under high pressure and high temperature",
Y. Katayama et. al., J. Synchrotron Rad. (1998), 5, 1023-1025

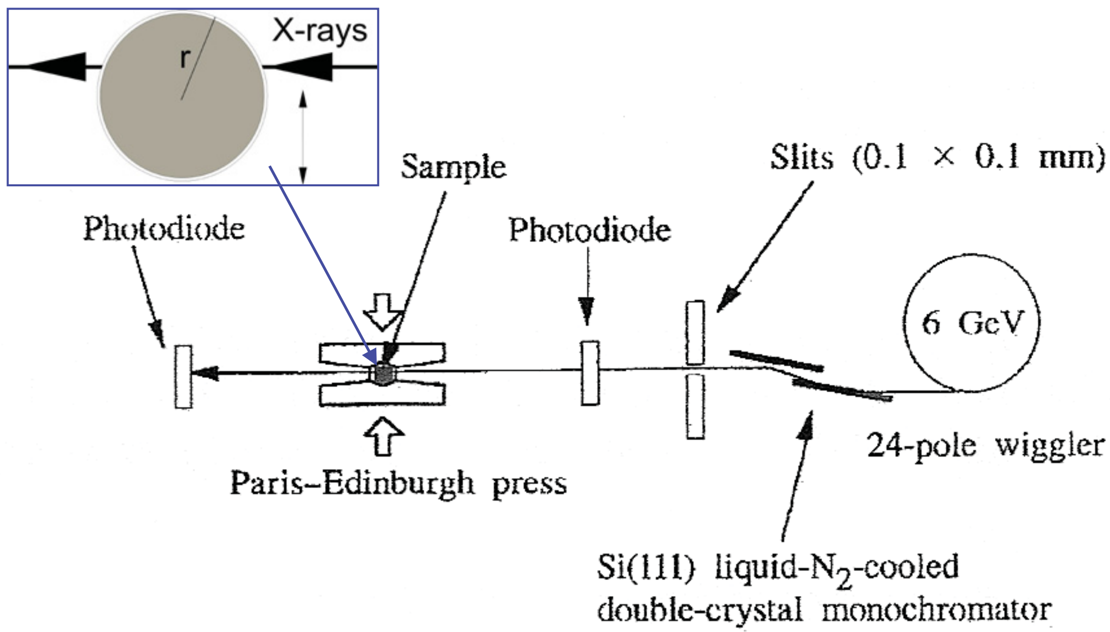


Figure 1
Schematic diagram of the experimental arrangements.

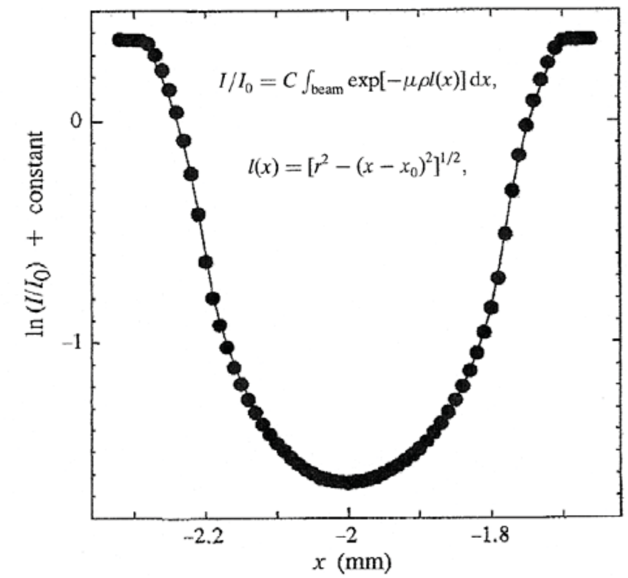


Figure 3
Logarithm of I/I_0 for liquid Bi at 1 GPa and at 750 K as a function of sample position. Circles are experimental data. The line is the result of parameter fitting.

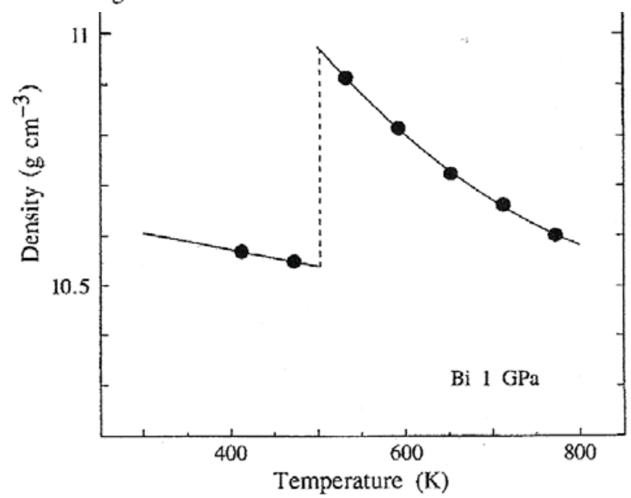
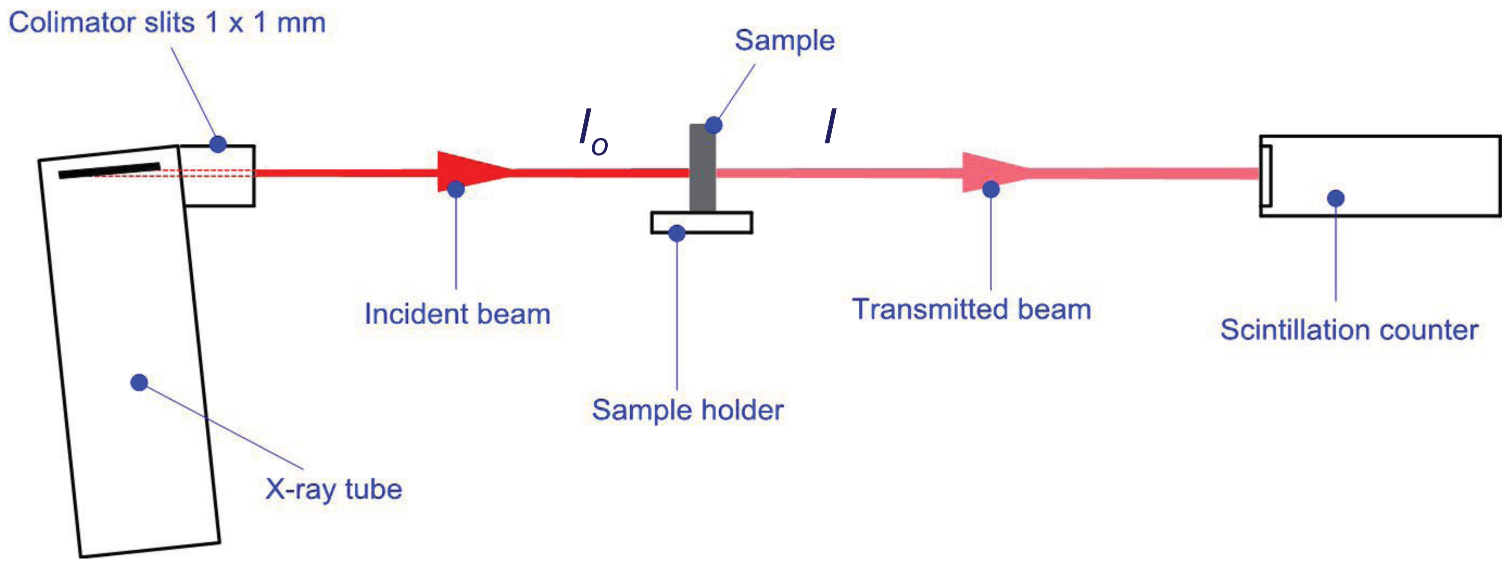


Figure 4
Density of crystalline and liquid Bi at 1 GPa as a function of temperature.

Y. Katayama et al.: *... the error was less than 1%...*

Available equipment



X-ray powder diffractometer (Seifert XRD 3000 PTS)

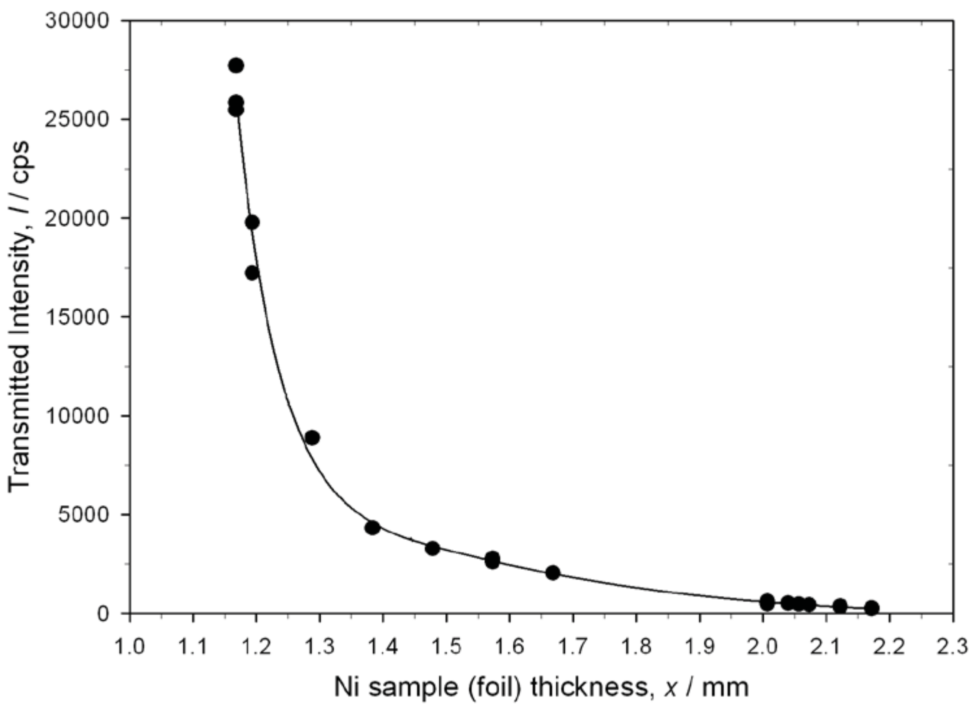
Tube: Cu fine focus, 1,5 KW

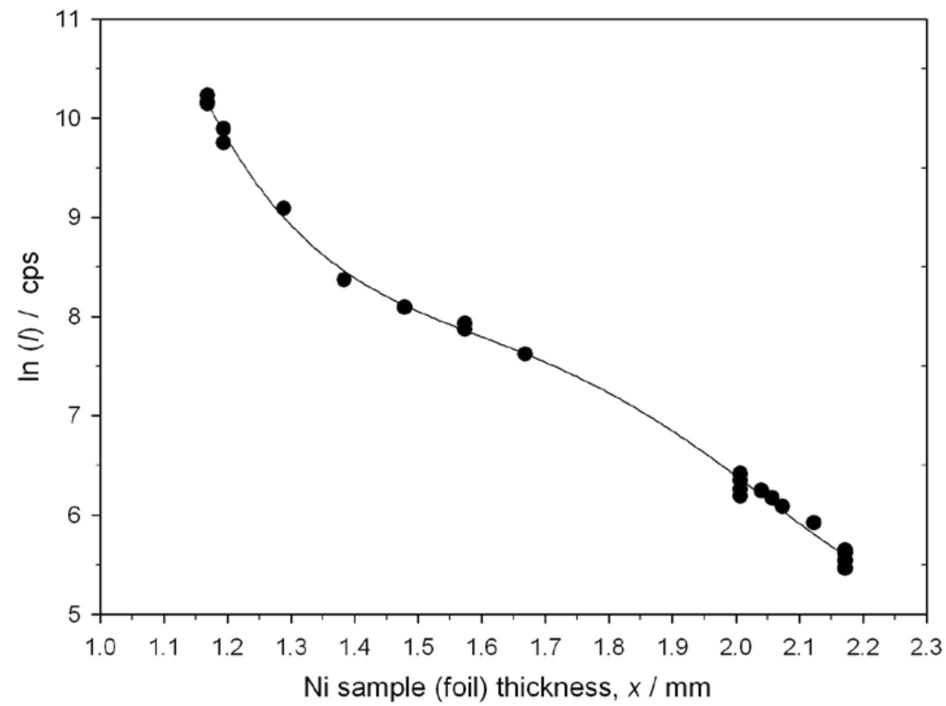
Wavelength: Ni filtered K_α (8.05 keV)

Collimated incident beam: 1 mm x 1 mm

Detector: NaI scintillation counter

Initial measurements

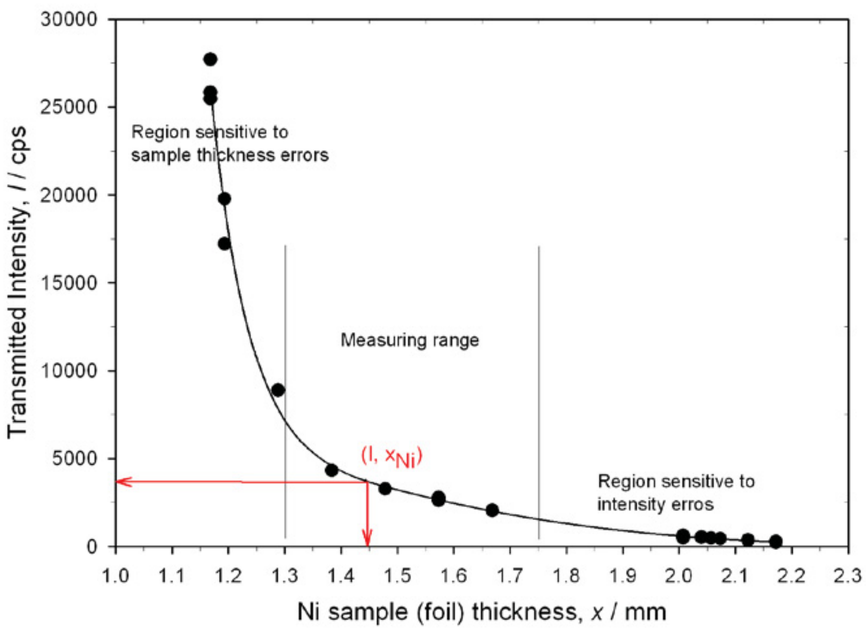




Intensity (I) of X-rays transmitted through Ni foils

Remark: Data deviate from attenuation law due to experiment conditions, mainly to detector’s specification

Preliminary results-1



For an "unknown" sample: $ln(I_o/I) = \rho_s \mu_{m,s} x_s$

In the graph, intensity I corresponds to Ni sample of thickness x_{Ni}, that is: $ln(I_o/I) = \rho_{Ni} \mu_{m,Ni} x_{Ni}$

Therefore: $\rho_s = (\rho_{Ni} \mu_{m,Ni} x_{Ni}) / (\mu_{m,s} x_s)$ (1)

If the transmitted intensity I is too high, Ni foils with thickness $x_{Ni,\ extra}$ can be placed in the front side of the sample, and Eq. (1) is modified:

$$\rho s = [\rho_{Ni} \mu_{m,Ni} (x_{Ni} - x_{Ni,extra})] / (\mu_{m,s} x_s) \qquad (2)$$

Preliminary results-2

Sample	*Thickness, x*	*μ_m for λ=8.05 keV*	*Known density, ρ*	*Density, ρ X-ray absorption*
Cu	*0.529 mm*	*51.37 (cm^2/g)*	*8.94 g/cm^3*	*8.90 g/cm^3*
Natural UO_2	*0.355 mm*	*269.49 (cm^2/g)*	*10.64 g/cm^3*	*10.62 g/cm^3*

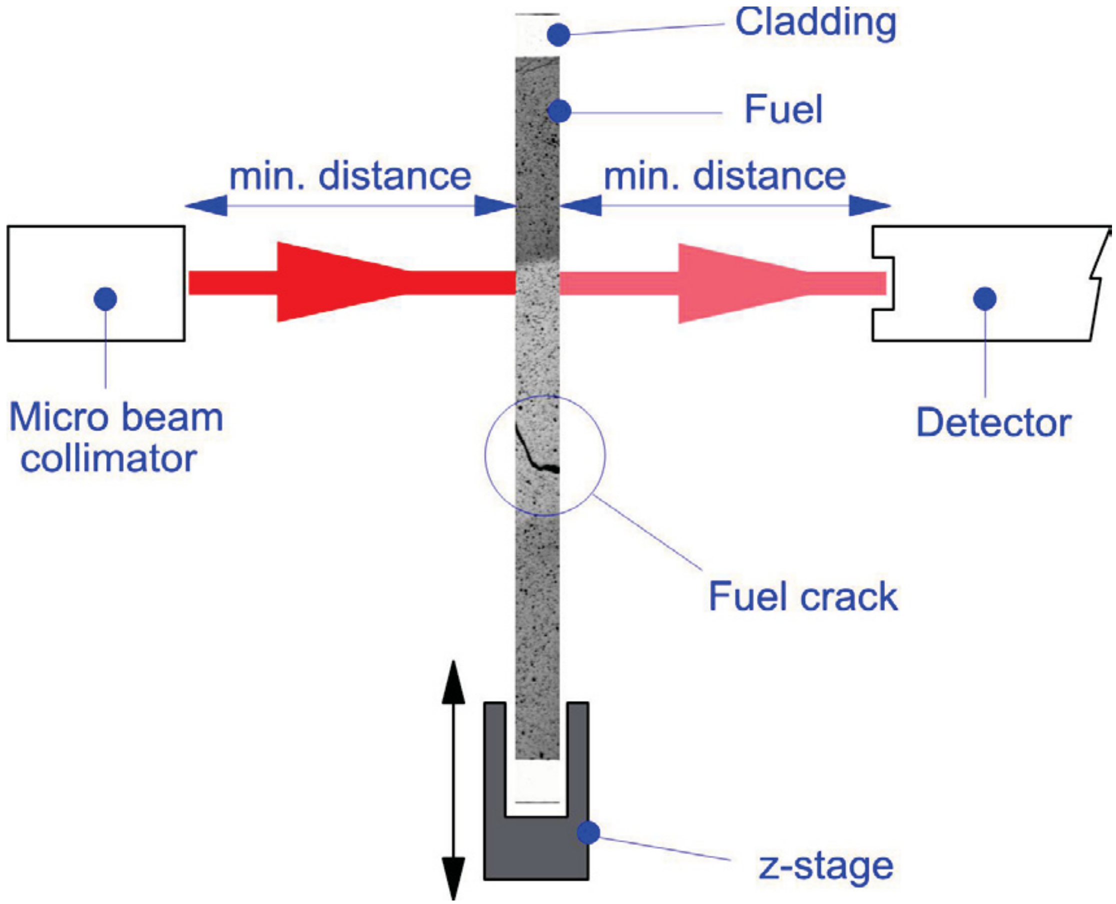
Notes:

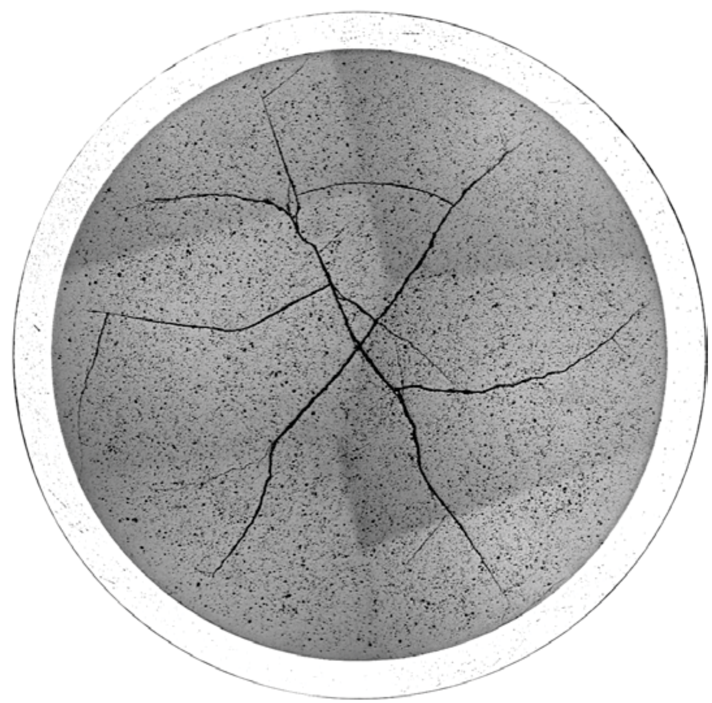
The Cu measurement is based on the Ni calibration graphs of last slides (99.9% pureness and ρ=8.899 g/cm^3)

The natural UO_2 measurement is based on calibration with depleted UO_2 disks and ρ=10.011 g/cm^3

Carry over to irradiated fuel demands

Optimum measuring conditions:





The X-ray absorption as complementary/alternative to immersion technique

Advantages

•non destructive method, a fuel disk with parallel sides is required

•not affected by temperature and pressure fluctuations in the hot cell

•radial total porosity/density profiles on fuel pellets can be measured

•Easily repeatable and fast measurements

Disadvantages

•fuel cracks must be avoided (similar to diffusivity measurements with laser flash)

•difficult calibration (solid fission products can be not considered, only empirical corrections)